ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

Prepared for DIVISION OF TECHNICAL INFORMATION / DIVISION OF ISOTOPES DEVELOPMENT, UNITED STATES ATOMIC ENERGY COMMISSION, by P. S. BAKER, A. F. RUPP, and ASSOCIATES, OAK RIDGE NATIONAL LABORATORY

Summer 1965

FEATURE ARTICLE:

Comparison of Electron-Beam and Gamma-Irradiation Plants

● VOLUME 2

● NUMBER 4

TECHNICAL PROGRESS REVIEWS

To meet the needs of industry for concise summaries of current atomic developments, the Atomic Energy Commission is publishing this series, Technical Progress Reviews. Issued quarterly, each of the reviews digests and evaluates the latest findings in a specific area of nuclear technology and science.

The five journals published in this series are:

Isotopes and Radiation Technology, P. S. Baker, A. F. Rupp, and associates, Oak Ridge National Laboratory

Nuclear Safety, Wm. B. Cottrell, W. H. Jordan, and associates, Oak Ridge National Laboratory

Power Reactor Technology, formerly prepared and edited by W. H. Zinn and J. R. Dietrich, Nuclear Division, Combustion Engineering, Inc. (a new contractor will take over preparation of this journal beginning with Vol. 8, No. 4).

Reactor Fuel Processing, Stephen Lawroski and associates, Chemical Engineering Division, Argonne National Laboratory

Reactor Materials, R. W. Dayton, E. M. Simons, and associates, Battelle Memorial Institute

Each journal may be purchased from the Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 20402. *Isotopes and Radiation Technology* at $2.00 per year for subscription or $0.55 for individual issues; the other four journals at $2.50 per year and $0.70 per issue. See back cover for remittance instructions and foreign postage requirements.

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A REVIEW OF RECENT DEVELOPMENTS

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SUMMER 1965

VOLUME 2

NUMBER 4

Foreword

Isotopes and Radiation Technology is a Technical Progress Review prepared by the Isotopes Development Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Power Development
Applications in Medicine
Safety
Miscellaneous Items of Interest

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FEATURE ARTICLE

Comparison of Electron-Beam and Gamma-Irradiation Plants

By Charles Artandi and Walton Van Winkle, Jr.*

Ethicon, Inc., has been using ionizing radiation for production-scale sterilization of sutures and other medical products since 1956. After years of experience with electron accelerators, the Company built a ^{60}Co radiation plant, which was put in operation in August 1964. Having the two types of plants on the same location for sterilization of the same products gave the opportunity for a thorough comparison. It was found that for sutures, pharmaceuticals, and disposable medical products, ^{60}Co sterilization offers several important advantages over the use of accelerators.

Introduction

The usefulness of ionizing radiation for sterilization, preservation, and chemical processing was demonstrated more than 15 years ago. In spite of the early glowing predictions by experts of imminent breakthroughs, progress in development of commercial irradiation processing has been fairly slow. This has not been due to lack of enthusiasm so much as to the absence of a well-coordinated program for developing these uses.

Accelerator manufacturers have been very active in selling their hardware. However, the chief customers have been government and institutional research laboratories, which have on their staffs highly trained technicians capable of keeping these machines in running condition for short periods of use. The needs for industrial applications have been few, and efforts to develop them have not been very rewarding. The absence of industrial radiation technology has inhibited both the seller and buyer from developing industrial processes using radiation.

Pioneering efforts in the development of industrial radiation sterilization with the use of accelerators was undertaken by Ethicon, Inc., and the story is well known by all who have an interest in this field.[1–3] Starting its research efforts with a 2-Mev Van de Graaff electron accelerator in 1953, Ethicon acquired a 3-Mev Van de Graaff and a 6-Mev microwave linear electron accelerator in 1957. The year 1956 marked the beginning of industrial radiation sterilization at Ethicon's Somerville plant. In 1959 another 6-Mev linear accelerator was installed. All these machines, built by High Voltage Engineering Corporation (HVEC), were the first of their kind, and Ethicon and HVEC shared the burden of pioneering. Because of the newness of these accelerators, many problems had to be worked out during production use, and the manufacturer cooperated at every step.

The development of industrial applications for radioactive isotopes was even less advanced in the fifties. At the time Ethicon made the decision to use accelerators, isotopes were also considered. The high price of ^{60}Co and the nonexistence of design experience and in-use technology forced a decision in favor of accelerators. In the United States, industrial experience and technology in the utilization of isotopic radiation sources for processing has lagged behind other countries, real progress having been made only in the last couple of years. The statutory authority and organization

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of the USAEC preclude the possibility of active government leadership in this field, and the national laboratories, although available for technical assistance, suffer from the same organizational handicap. The nuclear industry has been interested in doing expensive and extensive design studies and in building "deluxe" plants but has not been willing to underwrite the cost of developing isotope radiation technology.

It is not accidental that the British built the first demonstration and production ^{60}Co irradiation unit.[4] This was followed by construction of several industrial radiation plants: one in Australia, two in England, and one in Scotland. The first plant built in England is owned by Johnson's Ethical Plastics[5] and the one in Scotland, by Ethicon, Ltd.,[6] members of the Johnson & Johnson companies. The very active participation of the United Kingdom Atomic Energy Research Establishment in providing industry, at a modest charge, with design know-how and operating experience based on their studies and actual measurements has allowed the nuclear engineering industry in British Commonwealth countries to offer radiation plants at reasonable prices.

When in 1962 Ethicon reexamined its future radiation capacity needs, it looked into isotope availability again. American, Canadian, and British proposals were evaluated, but all American companies who participated submitted cost estimates substantially higher than the Canadian and British firms. This was due largely to lack of design and operating experience. The Atomic Energy of Canada Ltd., which incorporates in one company the functions of both cobalt producer and plant designer and manufacturer, is a good example of an organization with a properly constituted isotope development program. Ethicon contracted with AECL to design and build three cobalt radiation plants, and the largest one began operation Aug. 17, 1964, at Somerville, N. J. A second one of identical design but of lower initial throughput was put in operation in November 1964 in San Angelo, Tex. Both plants are fully automatic and require only intermittent supervision. The third one, a small batch irradiator, was built at Peterborough, Canada, and was commissioned in November 1964.

The advantages and disadvantages of the two types of irradiators are compared here with regard to source and accessory equipment.

Source

Penetration

The use of isotopes presents no penetration problems. Products of every size and shape may be processed, and many items can be irradiated in their final shipping cartons. The important factor is the freedom one has in the design and handling of the packages.

Accelerators are very suitable for irradiating thin plastic films, coated wires, and other materials for which penetration requirements are low; however, they are generally unsuitable for some radiation-sterilization processes. The minimum energy that permits the practical use of accelerators for that purpose is 6 Mev. Unfortunately, the maximum energy is limited to about 12 Mev since above that value the probability of electron and photon disintegration processes, which would result in induced radioactivity, increases strongly.

Radiation Efficiency

Radiation utilization or efficiency of absorption, i.e., the percentage of total radiation emission that is absorbed at the desired dose in the products in which we are interested, is very nearly the same for accelerators and the newer cobalt plants, namely, 20 to 30%.

A significant loss of radiation from a ^{60}Co source results from geometrical problems. Although the source emits radiation in all directions, there is a limit to how much product we can put around it. In most designs the product is very close to the source (6 to 8 in.), and it overlaps the source considerably in both horizontal and vertical directions. Thus the angles of lost radiation at the top, bottom, and ends of the source are decreased to acceptable levels. Depending on source design (flat, cylindrical, or multiple arrangements), 5 to 25% of the emission is absorbed in the cobalt itself. Because of the very penetrating nature of gamma photons, not all of them can be absorbed; some will go through the products and be absorbed in the cell walls. Multipass conveyor systems are designed to improve this situation, but, by the law of diminishing returns, the increased cost and complexity of such systems limit the number of passes. Radiation losses in the parts of the conveyor that hold the products and are exposed to radia-

tion are minimized by good design and the use of lightweight materials.

Accelerator radiation is characterized by the ionization absorption curve of the particular electron energy. Generally the entrance or surface dose is only 60 to 80% of the maximum, which is reached at about 30% of the total penetration. Of the total ionization, 27% is wasted as overdose since the desired radiation dose has to be set at the lowest (surface) dose. The tail end of the ionization curve (about 13%), which falls below the surface dose, cannot be utilized because any dose would be lower than desired. Cross-firing or splitting the beam may improve this but at a considerable increase in cost. Additional losses are due to the fact that the electron beam must be scanned wider than the products to be irradiated to ensure proper coverage. Consideration of all these losses shows that only 50 to 60% of the emission can be usefully absorbed. Products of uniform density and rectangular shape, e.g., plastic films, permit efficient use of radiation since here even some of the tail end of the ionization distribution curve may be used. However, most pharmaceutical and other products that have been processed by radiation are of irregular shape and nonuniform density. Some radiation will go through unused at points where there is not enough absorbing mass or where there is space between the products.

Plant Efficiency

Plant efficiency, defined as the percentage of time the plant is available for use when desired, differs significantly between the two types of facilities with respect to both capacity and maintenance cost. It bears no necessary relation to actual use because a plant may not be in operation for many reasons (weekends, holidays, scheduling, vacations, etc.). Most cobalt plants, because of their inherent simplicity, operate at 90% efficiency or better. Machine facilities are available 70 to 80% of the time, the remainder being spent in maintenance and repairs. Although availability may be higher for short periods, this figure is correct for long periods of operation.

Inventory

There is little difference in inventory costs for the two plants if processing time is within 24 hr. With cobalt irradiation, dose rates are low. The time required to deliver several megarads varies from hours to days, depending on the source size. To be effective, the source should be surrounded by a large amount of product during the various stages of irradiation. For items of high unit value, the processing time should be kept within a day; for other products a holdup of 2 to 3 days may be tolerated. With accelerator irradiation, on the other hand, dose rates are high. The absorption of several megarads takes only a second or so; therefore the products can be moved through the plant in small units at a fast rate. The total time from input to output may be only 5 to 10 min. For smooth operation and scheduling, it is advisable to have an in-process inventory buildup of ½ to 1 day's production ahead of the input.

Accessory Radiation Facilities

Besides the source, there are four essential parts of a radiation processing plant: shield, conveyors, controls, and safety systems.

Shield

A concrete cell of sufficient thickness (5 to 7 ft) is needed to absorb all radiation that is not absorbed in the products. Its purpose is to protect all personnel in the vicinity. Although the accelerator-produced electrons are much less penetrating than the gamma rays from a ^{60}Co source, these electrons, in turn, produce, through bremsstrahlung, high-energy X rays as by-products. Therefore shielding requirements for accelerators are similar to those for isotopic sources, and shield construction costs are about the same for the two plants, with possibly a slight bias in favor of accelerators.

When not in use, accelerators can be turned off, but isotopic sources must be shielded to allow access to the radiation cell. Two storage systems are in use for isotopic sources: dry and wet. Dry systems use air- or water-cooled concrete pits into which the source is lowered when not in use. A heavy metal plug on top of the source plaque or a movable one on the floor completes the shield. Wet systems (Fig. 1) use a stainless-steel-lined or unlined concrete pool 18 to 20 ft deep and filled with deionized and filtered water. Only for exceptionally large sources may cooling of the water be required.

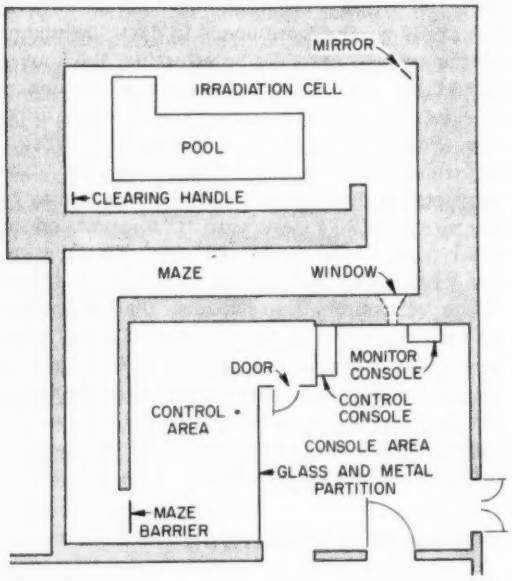

Fig. 1 Cobalt-60 irradiation facility.

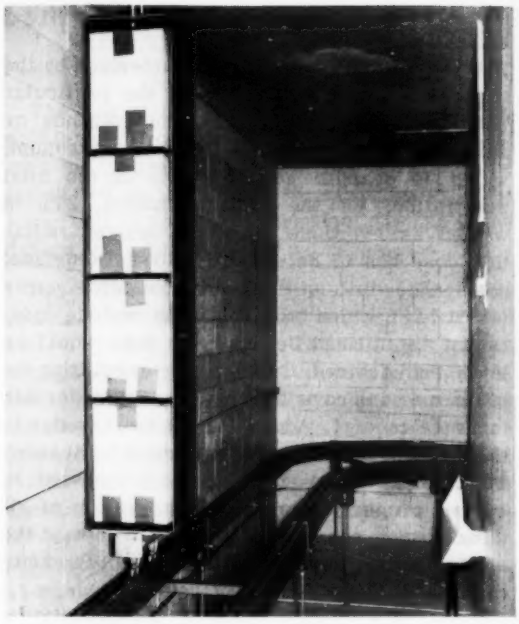

Fig. 2a Feed conveyor. A carrier with four product boxes is shown in the maze on its way into the cell.

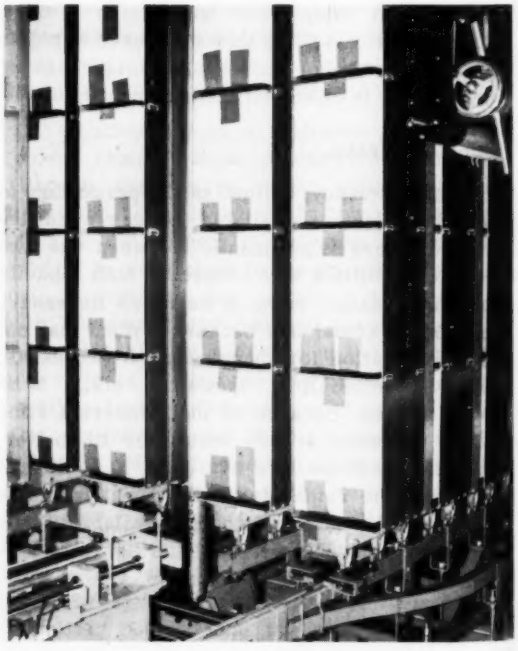

Fig. 2b Source conveyor showing four passes of product carriers. The carriers are back to back in each row.

Conveyors

It is fair to say that a conveyor system for a ^{60}Co plant is about twice as costly as that for an accelerator plant.

The conveyors for a cobalt plant are complex and are generally divided into two sections: feed conveyors and source conveyors (Figs. 2*a* and 2*b*). Feed conveyors are usually the power-chain type but may be the belt or push type. Their purpose is to move fresh products quickly into, and irradiated products quickly out of, the cell through the maze (or labyrinth).

Source conveyors are of either intermittent- or continuous-motion type. For the most efficient use of radiation, the products overlap the source in both directions (Fig. 3) and are carried around the source in two or more passes. The motion of the product around the source produces the effect of an "infinite source" by exposing all products in all positions to radiation and ensures uniform dose absorption in the direction of travel. Because of the low dose rate (only a few hundred rads per second) and the many positions in the various passes around the source, for all practical purposes it makes no difference with respect to dose uniformity whether the motion

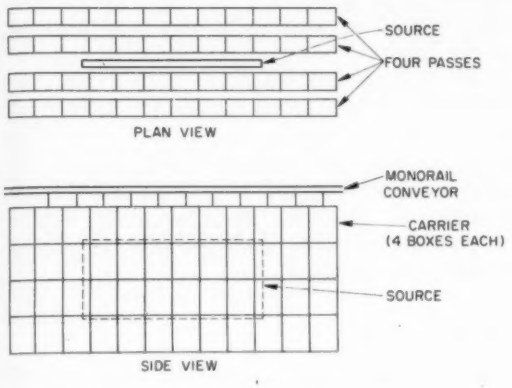

Fig. 3 Product overlap in relation to source size.

is continuous or intermittent. Intermittent source conveyors are operated hydraulically or pneumatically. Another mechanism is needed to make the dose uniform normal to the direction of travel. If the products are not moved perpendicularly to the direction of travel, the parts of the product that extend beyond the ends of the source absorb less radiation. The products are therefore shifted through all positions perpendicular to the motion of the source conveyor by a push or product-transfer mechanism. This may be located either inside or outside the cell.

In the case of accelerator irradiation, because of the limited penetration of the electrons (6 Mev is equivalent to 0.8 in. useful penetration of water), products must be carried in thin layers on trays to ensure that a proper dose is delivered throughout. Conveyors must be of the belt type (rubber outside the radiation cell, stainless steel inside) and are relatively simple since the product makes only one pass at one level (Fig. 4). Many turns are required to negotiate the maze in a small cell, but the number may be decreased by building a larger cell. The trouble-free operation thus

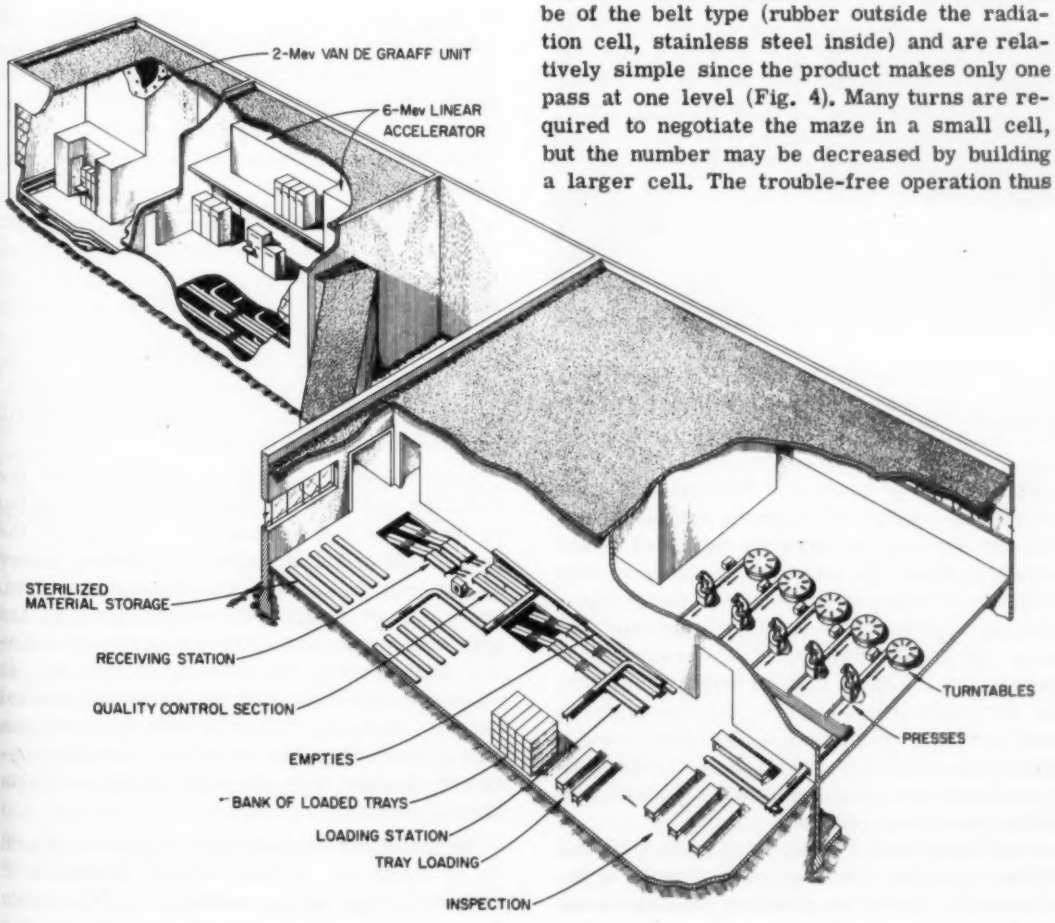

Fig. 4 Accelerator facility.

achieved pays for the extra expenditure in a short time. Because the dose rate is very high (up to several megarads per second), speed control is of extreme importance. Therefore a good variable-speed conveyor section under the accelerator radiation source is needed for both control and necessary adjustments of speed to ensure proper dose under all operating conditions.

Controls

The control systems of ^{60}Co plants (Fig. 5) are considerably less costly than those of accelerator plants.

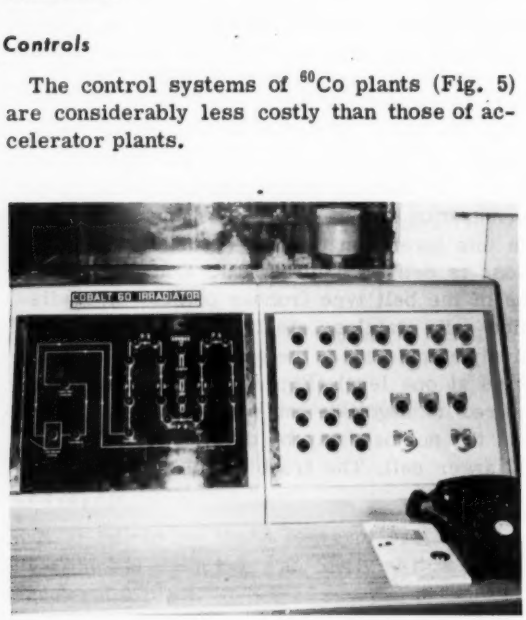

Fig. 5 Control console of ^{60}Co plant. Indicator panel gives instantaneous view of operation. Control and maintenance panel is on the right.

The electronic part of a ^{60}Co plant is quite simple; the mechanical systems are automated, and the whole plant requires only intermittent attention. When the source is exposed, it emits radiation of known energy and adequate penetration in a geometrical configuration that has been previously defined. The decay rate is known, and the dose is determined at any time by the total cobalt charge and the exposure time (or conveyor speed). The dose rate is about four orders of magnitude lower than that of accelerators; therefore conveyor-speed control is much less critical.

In the accelerator plant, in addition to control of the complex electronic components of the accelerator itself, the following variables must be carefully controlled for reliable radiation processing: electron-beam energy (Mev), beam current, scan width, and conveyor speed. Energy control is extremely important because of the inherent penetration limitation and because changes will affect the scan width. The beam current may affect the energy. Control of scan width is essential for complete coverage of the product, and conveyor speed is a critical factor because of the very high dose rates produced by machines. Most machines require the constant attention of an operator for making adjustments as they become necessary.

Safety Systems

Two of the really important parts of any radiation processing safety system are the shield, discussed above, and properly trained personnel. Thorough training of operators; existence of a clearly written, up-to-date, and detailed operating procedure; and enforcement of adherence to the procedure are essential for safe operation. An intricate system of interlocks, devices, and measuring instruments enhances safe operation. The interlock and monitoring systems are more costly for a ^{60}Co plant, but these systems are a relatively small part of the total cost.

Most radiation plants have a viewing system to allow the operator to scan the radiation cell before startup. Before operation starts, all safety interlocks (doors, barriers, etc.) must be closed. The operator then locks the outside entrance to the cell (the last interlock) with the only key, which is also used to start the radiation plant. If any interlock is open, nothing will happen when the key is turned.

Some plants enforce an inspection of the radiation cell by the operator before startup by requiring the pulling of a handle or the pressing of a button in the most distant corner of the cell to actuate the interlock system. Other plants incorporate safety mats at the entrance which shut down operations if someone inadvertently tries to enter the cell or utilize, in the floor past the entrance, physical barriers that are closed or pits that are open during operation. Any startup of radiation operation begins with the sounding of an alarm to warn people.

Accelerator facilities may be entered as soon as a shutdown occurs because emission of radiation stops instantaneously. In ^{60}Co plants the source has to be lowered into a pit or a pool of water. The time required is generally

less than 2 min, but a radiation hazard exists until the source reaches the "safe" position.

After installation of either type of radiation plant, an extensive survey is made to measure radiation levels at the various locations outside the cell in restricted or nonrestricted areas. State and federal regulations specify maximum permissible radiation levels for the various areas. If everything is right at that time, certain factors have to be watched to ensure that conditions do not change. In an accelerator installation:

1. The energy and beam current must not exceed the maximum values used in the checkout.

2. Proper focusing of the beam must be maintained to prevent powerful X rays from being produced by impingement of high-energy electrons on metal parts inside the accelerator tube.

3. Good shielding of power-generator tubes must be maintained. The background radiation must be monitored carefully, and all the walls around the facility must be surveyed periodically.

In cobalt plants:

1. Cobalt must not be added without a new radiation survey.

2. The integrity of the source must not be impaired. Radioactive cobalt metal is generally doubly encapsulated for protection against corrosion, and the first indication of damage would be the appearance of cobalt oxide dust in the air or, in case of wet storage, particles suspended in water.

3. Cobalt must not be inadvertently removed from the source frame.

Properly located radiation-monitoring equipment ensures safety of operations. Separate radiation monitors are set up to measure radiation levels at the following locations: (1) the cell when the source is in "safe" position; (2) inside the entrance door to ensure that no cobalt is carried out; (3) background of the control area; (4) air filter; and (5) water deionizer in case of pool storage. Periodic radiation surveys are not needed since there can be no change in the emission except for gradual decay (about 12% per year).

In addition to all these safety precautions, in both plants all operators wear film badges and undergo periodic medical checkups.

Cost Comparison

Capital Investment

A 6-Mev linear accelerator of about 5 kw output requires a capital outlay of about $200,000, i.e., $40,000 per kilowatt. One kilowatt equivalent of cobalt radiation costs about $45,000. It is important to point out that, although accelerators can be turned off with no wear on the machines, ^{60}Co decays at a constant rate regardless of whether it is used or not. However, both types of installations are amortized on a time basis regardless of the amount of use for processing; therefore the calculated operating cost takes care of this problem.

Another important consideration is that of increasing radiation capacity in case of need. The capacity of a machine radiation plant can be increased above its practical maximum only by adding another unit with its shield, conveyors, controls, etc.—a complete new installation. If enough foresight is exercised during the design stage of a cobalt plant, its capacity may be significantly increased by the simple addition of cobalt, assuming that the shield is thick enough and the source frame is large enough. Then only the conveyors have to be speeded up to increase the throughput. No new capital investment is required; only cobalt need be bought.

Operation and Maintenance Labor Costs

The total of salaries, overtime payments, and indirect labor charges for ^{60}Co plants is only 20 to 25% of that for accelerator plants. Maintenance costs for ^{60}Co plants are estimated to be about 10% of those of accelerator plants because of the vulnerability of the latter to breakdowns and the need for replacement of costly parts.

The manpower requirement for ^{60}Co plants is very low because of their simplicity, much greater reliability, and capacity to irradiate products in much larger units. Under the intermittent supervision of a professional person, four technicians can operate the plant three shifts a day, five days a week. The same technicians can handle sampling, dosimetry control, and unloading of irradiated products. Only one wage person is needed per shift to load the input conveyor and keep records.

For operating an accelerator facility two shifts a day, five days a week, a full-time

manager, an assistant manager—both with a background of electronics engineering—and four or five technicians are needed. The extra technicians are required to do the necessary repair and preventive maintenance work and to cover the work load in excess of two shifts. The manager and assistant manager are responsible for direction, troubleshooting, and planned improvements in equipment and controls. Quality-control personnel are needed to keep records of materials that go through the process and to sample the batches for both sterility and physical testing. They also perform dosimetry work to check scan width, scan uniformity, penetration, and radiation dose. Because of the limited penetration of accelerator electrons, products to be irradiated have to be handled in small units (trays). Therefore two wage persons are required to man the input and output ends of the conveyor system, to prepare materials for irradiation, and to remove irradiated products. Quality-control and wage personnel must be available whenever the accelerators are in operation regardless of the time of day or week. This results in a high level of overtime payments and associated premiums.

Utilities

Utility costs for the two plants are similar but are a little more favorable for ^{60}Co plants.

Depreciation

A great deal depends on the original cost of the installation and on the rate of depreciation. If there is only one accelerator in the facility, depreciation charges for the two types are similar. However, two accelerators are usually required, partly to have one as a standby and partly to have some reserve capacity in case of uneven product flow. In this case, depreciation charges of an accelerator plant will be appropriately higher.

Processing Costs

As a result of the above-discussed factors, the processing cost on a weight or unit basis is two to three times higher for accelerator plants than for ^{60}Co plants:

	^{60}Co	Accelerators
Labor costs, operation	1	4–5
Labor costs, maintenance	1	10
Utilities	1	1
Depreciation	1	1–1.5
Processing costs	1	2–3

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Section

I

Isotopes and Radiation Technology

Isotope Technology Development

In this section papers on activation analysis from the Third International Conference on the Peaceful Uses of Atomic Energy are presented. These papers include a review of recent advances in neutron activation analysis, neutron counting in activation analysis, and two papers on the application of activation analysis to the specific problems of analysis of trace elements in lake waters and in metals such as lead and aluminum.

The Geneva Conference papers in this section and in Sec. II, Safety, are handled in the same way as were those Geneva papers which were in the previous issue of *Isotopes and Radiation Technology*.* Only significant technology concerning radioisotopes is given, and references that could not be checked are marked with an asterisk.

Advances in Neutron Activation Analysis

By Vincent P. Guinn†

Paper 197

During the past six years, neutron activation analysis has advanced rapidly in the United States. New and better equipment and techniques have increased its capabilities and areas of useful application. The widespread use of the small neutron generator, coupled with the purely instrumental form of the method, is enabling activation analysis to be employed widely and effectively. It now competes with other methods of elemental analysis at both the minor- and major-constituent levels of elements although previously it was largely restricted to trace-level determinations. The method has also been improved at trace-level concentrations by the use of high-flux research reactors with special provisions for efficient neutron activation, high-intensity reactor pulses, and fast reactor neutrons. The principal improvements in equipment, techniques, and applications in the physical sciences are discussed below.

*See *Isotopes and Radiation Technology*, 2(3): 199 (Spring 1965) for a complete description of the type of coverage given Geneva papers.

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Improvements in Equipment

Marked advances have been made in the performance and availability of various types of neutron sources for activation analysis and in counting equipment used for detection of the induced activities.

NEUTRON SOURCES

The low-intensity isotopic neutron source, which is useful essentially only for teaching purposes and some analyses for high-sensitivity elements at high levels, has been improved by the availability of ^{241}Am-Be sources. These have the desirable high neutron flux of the earlier ^{210}Po-Be and ^{239}Pu-Be sources but do not have the undesirable short half-life of the ^{210}Po sources (138 days). Americium-241 has a half-life of 458 years, which gives it a considerably greater specific activity than that of 24,360-year ^{239}Pu. Americium-beryllium sources capable of producing up to 10^8 n/sec are now commercially available. The hoped-for ^{252}Cf spontaneous-fission neutron source is still not commercially available but should be in the foreseeable future. Strain[1] and Strain and Leddicotte[2] reported the use of americium-beryllium sources for activation analysis, and papers on isotopic sources were published recently by Kinsey et al.,[3] Kohn and Green,[3] and McFarling et al.[3] Similar

neutron-output rates are possible with small sealed-tube accelerators, such as those described by Oshry.[4]

Small-Accelerator Neutron Sources. Development of these sources has advanced rapidly in recent years. The most widely used accelerator for neutron irradiation is a small Cockcroft-Walton machine that produces 14-Mev neutrons by bombardment of a tritium target with 150-kev deuterons. With a D^+ beam current of 1000 μa, a 5 curie/sq in. tritium target emits, isotropically, about 10^{11} n/sec. At positions where samples of appreciable size (1 to 10 g) can be placed and irradiated without too large a flux gradient, 14-Mev neutron fluxes of about 10^9 n/(cm^2)(sec) can be obtained. More modest versions of such accelerators (100 kev, 200-μa current) produce about 10^{10} n/sec and cost appreciably less. Properly installed for neutron activation analysis, the 10^{11} n/sec accelerator requires approximately 6 ft of concrete shielding (or its equivalent), plus 10 to 15 ft of distance, for safe continuous use. Somewhat less shielding is needed at a 10^{10} n/sec rate.

The principal limiting problem at present is the rapid deterioration of the tritium targets with use at maximum beam power. A typical target is a thin circular copper sheet, about 1 in. in diameter, which has on one face a few-micron-thick layer of partially hydrided (by exposure to tritium gas) titanium containing approximately 5 curies of tritium per square inch. Since a 150-kev D^+ ion penetrates only about a 1-μ depth of the titanium tritide layer, at full power 150 watts of thermal energy is liberated in this thin layer. Even with the beam defocused so that it is spread over an area of about 1 sq in. and with water cooling on the back of the target, the tritide layer is heated considerably during full-power operation. This results in decomposition of the metal tritide and loss of tritium to the vacuum system of the accelerator with an appreciable decline in the target output rate. Typically, at 150 kev and 1000 μa, output will decrease by a factor of 2 in about 30 min of operation with a target of 1 curie/sq in. and in 1.5 hr with a target of 5 curies/sq in. The output rates of both targets continue to decline with further use but not so rapidly. If a reasonably high neutron flux is to be maintained steadily for many hours, it is best to start with a fresh target at a moderate beam current, perhaps 300 μa, and gradually increase the current to compensate for target decline. This can be done automatically, if desired, by installing a servomechanism that gradually increases the beam current to maintain a constant neutron-flux monitor reading. At lower beam powers the decline rate is not so rapid, varying, with a fresh target, approximately with the inverse square of the beam power. One commercially available accelerator now has a useful maximum beam current of 2.5 ma, which provides more reserve power for increasing beam current and thus extends the time between target changes.

There have been efforts to improve target lifetimes. The older oil-diffusion pump has been largely replaced by the Vaclon pump to prevent deposition of material from the pump oil onto the surface of the target. Freon cooling of the target is available instead of water cooling and yields some improvement in target lifetime. The 5 curies/sq in. target provides both an appreciably higher initial output rate and a lower decline rate than the older 1 curie/sq in. target, but it is somewhat more expensive. Larger rotating targets are now available. These provide lifetimes of 5 to 10 normal targets without the necessity of frequent shutdowns for target replacement. Further defocusing of the beam is helpful to target life, but some loss in neutron flux occurs at sample locations. Yttrium tritide targets have been prepared and tested and should be less susceptible to decline from thermal decomposition since yttrium hydride is thermally more stable than titanium and zirconium hydrides. So far, however, it appears that improvements in target lifetime achieved with yttrium tritide have been rather small. One commercial accelerator now incorporates a target-reloading system, which permits a partially spent tritium target to be periodically recharged by bombarding it for a time with T^+ ions at a reduced accelerating potential. This procedure essentially restores a partially spent target to its original output level and can be repeated several times. Target lifetime is also lengthened by magnetically deflecting D_2^+ ions present in the D^+ beam away from the target. The D_2^+ ions cause target heating, as do the D^+ ions, but contribute very little to neutron production. With accelerators using a radio-frequency ion source, the D_2^+ ions cause little trouble since the beam from such a source is about 90% D^+ and only 10% D_2^+. However, these heavier ions are a serious problem with the less expensive, simpler, and longer lived

Penning type ion source, which produces a beam 90% in D_2^+ ions and only about 10% in D^+ ions. Such sources currently do not deliver as large a D^+ ion current as do the radio-frequency sources.

For thermal-neutron activation with a moderate-cost accelerator, a 2-Mev positive-ion Van de Graaff has advantages over the smaller, less expensive Cockcroft-Walton accelerator. It generates 10^8 to 10^9 n/(cm^2)(sec) thermal-neutron fluxes by the ^{9}Be$(d,n)^{10}$B reaction; thus there is no problem of target lifetime.

Neutron generators and their uses have been described by Anders,[5] Burrill and MacGregor,[6] Gillespie and Hill,[7] Guinn,[4,8–10] Guinn and Steele,[11] Meinke,[12] Meinke and Shideler,[13] Mott et al.,[14] and Strain.[1]

Nuclear Research Reactors. The pool reactor is the type most widely used for neutron-activation-analysis studies and is generally operated at power levels from 100 kw to a few megawatts. These reactors provide neutron fluxes from 10^{12} to 10^{14} n/(cm^2)(sec), depending on the type, power level, and location within the pool or core. These simple below-ground reactors are moderate in cost compared with earlier models, and many are being used in the United States for neutron activation analysis. Fewer water-boiler and graphite-lattice research reactors are now used in the United States for such work. The first reactor ever used for neutron-activation-analysis studies, the Graphite Reactor of Oak Ridge National Laboratory, was finally shut down in November 1963 on its twentieth anniversary.

One of the newer types of pool reactors uses uranium–zirconium hydride fuel elements. This is the TRIGA reactor, which is used very widely in the United States and abroad. The hydride fuel element provides two desirable features: intrinsic safety and the ability to pulse to very high flux levels. The 250-kw TRIGA Mark I reactor at General Atomic (GA) provides thermal-neutron fluxes of 2 to 4×10^{12} n/(cm^2)(sec) in regularly available irradiation positions and 10^3-watt pulses that provide peak neutron fluxes of about 4×10^{16} n/(cm^2)(sec). Buchanan[4,15] tabulated the detection sensitivities attainable for 69 elements with a 1.8×10^{12} n/(cm^2)(sec) thermal-neutron flux and 1-hr irradiation in the 250-kw TRIGA reactor. These sensitivities range from 10^{-8} to 50 μg with a median of 0.01 μg. Meinke[12] tabulated sensitivities for a 10^{12} n/(cm^2)(sec) flux and various irradiation times. The 1000-kw TRIGA, which has four times the wattage of the 250-kw TRIGA, provides steady fluxes of 10^{13} n/(cm^2)(sec) and peak fluxes, in a pulse, of about 10^{17} n/(cm^2)(sec). This reactor has a rotary specimen rack that makes it especially effective for neutron activation analysis. The rack contains 40 tubes and rotates around the core during irradiation; therefore the average neutron flux at a given height is exactly the same for all 40 locations. Stacking as many as 6 samples vertically in each tube provides a total capacity of 240 samples for a single irradiation, each group of 40 having the same flux. The vertical flux gradient in the 40 tubes is not large and is accurately known. Research reactors and their applications to neutron-activation-analysis studies have been described by Buchanan (Refs. 4 and 15), Guinn (Refs. 4, 10, and 16–19), Meinke (Refs. 20–22), and Schmitt (Ref. 23).

MULTICHANNEL PULSE-HEIGHT ANALYZERS AND COUNTERS

In recent years the vacuum-tube multichannel pulse-height analyzer has been largely superseded by completely transistorized units. They are more reliable, smaller, lighter, and more versatile and consume less power than their vacuum-tube predecessors. Typically, the modern type incorporates 200 to 512 analysis and storage channels (with provisions for dividing the memory into two or four equal groups of channels); live and static oscilloscope display (linear or logarithmic); preset live time and clock time; a percentage-dead-time meter; digital readout (tape printer or electric typewriter); analog readout (oscilloscope camera, x-y plotter, or strip-chart recorder); and magnetic or punched paper tape for spectrum-stripping operations. A multichannel instrument can be used to analyze simultaneously the outputs of two or four scintillation counters. The only serious limitation is the relatively long, pulse-size-dependent dead time (typically amounting to $40 + 0.2N$ μsec, where N is the channel number) resulting from the use of the amplitude-to-time conversion principle rather than of the faster (but drift-prone) stacked-discriminator principle of the earliest multichannel analyzers.

The trend in detection equipment has been to the use of relatively large NaI(Tl) crystals directly coupled to a photomultiplier tube of comparable size. Backscatter is minimized by mounting the crystal in a 4-in.-thick lead shield

lined with thin sheets of cadmium and copper to minimize lead X-ray contributions to the observed spectrum. Crystal-photomultiplier combinations selected for maximum resolution and minimum gain shift at high counting rates are available and are very useful. Two commonly used crystals are the 3- by 3-in. solid type and the 5- by 5-in. well type. Fite et al.[4] incorporated a small alpha-particle source in the crystal and used its high-energy peak in the pulse-height spectrum with an electronic servomechanism to maintain a constant gain. A commercially available circuit is based on the same principle but has a high-energy peak rather than an alpha-particle peak in the sample spectrum for monitoring. In most gamma spectrometry a 1-cm-thick layer of plastic is placed between the sample and the crystal to screen out beta particles with minimum production of bremsstrahlung.

Multichannel gamma spectrometers and their uses in neutron activation analysis have been described by Brauer and Connally (Ref. 24), Crouch and Heath (Ref. 25), Guinn (Refs. 2 and 4), Heath (Refs. 4, 10, 24, and 25), Perkins (Refs. 4 and 10), and Steele et al. (Ref. 10). Heath[26] and Anders[27] published very useful gamma-spectrum catalogs. Strominger et al.,[28] in their Table of Isotopes, summarized the best values of half-lives, beta- and gamma-ray energies, and decay schemes; and Crouthamel[29] edited a book entitled *Applied Gamma-Ray Spectrometry*.

In beta counting and spectrometry, large well type plastic scintillators have found use. Semiconductor detectors are being applied in beta, X-ray, and low-energy gamma-ray spectrometry and have excellent resolution. The lithium-drifted detector provides relatively thick sensitive zones (up to 10 mm). The more dense germanium type gives better absorption than the silicon type but must be refrigerated. Still higher atomic number semiconductor detectors are under development. Borkowski[10] recently summarized the field of semiconductor detectors. Commercial low-background (0.2 count/min) beta counters are now widely used.

Advances in Techniques

Several new techniques for neutron activation analysis that have been developed within the past several years are summarized below.

USE OF REACTOR PULSES

High-intensity short-duration pulses attainable with a pulsed reactor can be used to improve the sensitivity of detection of elements that form very-short-lived induced activities. For example, in the GA reactor a single pulse produces approximately 500 times as much activity of a nuclide having a half-life of 0.1 sec as can be produced by operation at the normal steady-state power level, 50 times as much of a 1-sec half-life isotope, and 5 times as much of a 10-sec activity. For an induced activity with a half-life of 50 sec, the pulse offers no advantage, and for longer lived species it is progressively less sensitive than longer irradiations at the normal power level. Enhancement of sensitivities by means of pulsing has been demonstrated experimentally with induced activites from 7.35-sec ^{16}N, 12.4-sec ^{34}P, 4.8-sec ^{79m}Br, 16.1-sec ^{89m}Y, 7.2-sec ^{197m}Au, 10.7-sec ^{20}F, 5.5-sec ^{183m}W, and 19.5-sec ^{46m}Sc. Both thermal-neutron and fast-neutron induced activities of short half-life are enhanced by use of the reactor pulsing technique. The use of reactor pulses for neutron-activation-analysis studies has been described by Guinn[4,16,17] and Lukens et al.[10]

USE OF FAST REACTOR NEUTRONS

The great bulk of reactor-neutron activation analysis to date has been done with thermal neutrons. Little effort has been devoted to the effective use of the higher energy fission-spectrum neutrons that also are present in reactor cores at high fluxes (one well-known exception is the $^{32}S(n,p)^{32}P$ reaction for detection of sulfur). A study of activation analysis with fast reactor neutrons in the GA laboratory has shown that several elements (e.g., N, O, Si, P, Fe, Y, and Pb) can be detected with greater sensitivity by activation with fast neutrons than with thermal neutrons in the same reactor. Furthermore, many other elements are quite sensitively detected by activation with fast neutrons, though not so sensitively as with thermal neutrons in the same reactor (e.g., F, Mg, Al, S, K, Ti, Cr, Co, Ni, Zn, Ge, Se, Br, Sn, and Nb). Because of the additional choices of gamma energies and half-lives available, the use of fast-neutron rather than thermal-neutron-induced activity can decrease interferences from other activities.

In reactor irradiations the problem of minimizing (n,γ)-induced activities in the sample

matrix is serious if the objective is to detect an (n,p), (n,α), $(n,2n)$, or (n,n') product. Work to date with ^{10}B-lined tubes for suppression of (n,γ) activation has decreased the (n,γ) activation by factors of 100 to 200, depending on the element involved. Additional shielding with one or more elements that have large (n,γ) resonances may improve the (n,γ) suppression further. The use of ^{6}LiD or ^{235}U liners may be helpful not only in screening out low-energy neutrons but also in generating additional fast neutrons (14-Mev and fission-spectrum neutrons, respectively).

The fast-neutron sensitivities attainable in the GA reactor are 10 to 100 or more times better than those attainable with a small 14-Mev neutron generator, the ratio depending on the reaction threshold and the shape of the curve of cross section vs. neutron energy. In the pneumatic-tube position (outermost fuel-element ring) of the 250-kw TRIGA reactor where the thermal-neutron flux is 4.3×10^{12} n/(cm^2)(sec), the neutron flux above 1.35 Mev is 7.5×10^{11}, that above 3.68 Mev is 1.25×10^{11}, and that above 6.1 Mev is 1.9×10^{10} n/(cm^2)(sec), respectively. Fluxes of 14-Mev neutrons attainable with the small neutron generators are 10^8 to 10^9 n/(cm^2)(sec).

The use of fast reactor neutrons for activation-analysis studies has been described by Guinn (Refs. 4, 16–18, and 30), Meinke (Refs. 20 and 21), and Yule et al. (Ref. 10).

FAST RADIOCHEMICAL SEPARATIONS

Where interferences are very severe and cannot be sufficiently minimized by proper choice of irradiation and decay times, post-irradiation radiochemical separations with carriers and holdback carriers may be used. This is a long-established technique but has ordinarily been used with conventional chemical-separation methods. These are time consuming and hence preclude the effective use of induced activities with short half-lives. Meinke[4,12,21] pioneered the development of very fast radiochemical-separation techniques, in many cases achieving separations in 5 to 10 min. In the GA laboratory[10,16] these techniques are now regularly used successfully for radiochemical separation of such short-lived species as 2.3-min ^{28}Al, 3.8-min ^{52}V, 5.8-min ^{51}Ti, 8.7-min ^{49}Ca, 9.5-min ^{27}Mg, 10.5-min ^{60m}Co, and 25-min ^{128}I. The method is thus applicable to analyses formerly considered to be impractical by neutron activation because of severe interferences. DeVoe et al.[31] and Ruch et al.[32] used amalgam-exchange reactions in radiochemical separations. The National Research Council Nuclear Science Monograph Series gives radiochemical-separation procedures for many elements.[33]

PURELY INSTRUMENTAL TECHNIQUES

With the advent of the small neutron generator and the multichannel pulse-height analyzer, neutron activation analysis could be effectively applied at both minor- and major-constituent levels in competition with the many other methods of elemental analysis widely used at these higher concentration levels. This possibility arose because neutron activation could be much faster than other methods of analysis for many types of samples. Since the small generator was much less expensive than a nuclear reactor, equipment cost was not prohibitive. It was also clear that this method could compete with other methods only if purely instrumental techniques were employed; if radiochemical separations were also used, the great advantage of speed was lost. On this basis, Guinn and Wagner[34] developed a comprehensive system of purely instrumental thermal-neutron activation analysis using a 10^8 n/(cm^2)(sec) accelerator (3-Mev-electron Van de Graaff) flux and multichannel gamma-ray spectrometry. This method has been in routine use for the past 7 years, competing effectively with other analytical methods for the determination of approximately 25 elements. A small Cockcroft-Walton 14-Mev-neutron generator for routine fast-neutron activation analysis at fluxes in the 10^8 to 10^9 n/(cm^2)(sec) range augments the method. Many elements can thus be detected, by either thermal or 14-Mev neutrons, down to levels as low as 1 to 100 ppm in 10-g samples. This development was greatly aided by the earlier, but more limited, single-channel-analyzer work of Morrison and Cosgrove,[35,36] Cosgrove and Morrison,[37] and Cosgrove et al.,[38] who used reactor activation, and of Atchison and Beamer,[39] who used accelerator activation. Other aspects of this work have been described by Guinn,[4] Guinn et al.,[40] Wagner et al.,[41] and Johnson.[10] Many laboratories in the United States, particularly in the petroleum and chemical industries, have installed systems for purely instrumental neutron activation analysis in which small neutron generators are used. Interesting results

have been reported by Anders,[10,42–44] Burns,[10] Gilmore and Hull,[4,45] Gray and Cartwright,[10] Guinn,[8,9,46] Guinn and Johnson,[47] Iddings,[10] Mott and Rhodes,[10] Steele,[2] Steele et al.,[10] Steele and Meinke,[4,48] and Veal and Cook.[49]

The instrumental technique used with reactor activation has been described by Buchanan (Ref. 4), Guinn (Refs. 4, 10, and 16–19), Kramer and Wahl (Ref. 10), and Meinke (Refs. 20, 21, and 50).

SPECTRUM STRIPPING

The technique of simplifying complex gamma-ray pulse-height spectrums by means of spectrum stripping was first described by Lee,[51] although not for activation analysis. He used "live" stripping, i.e., counting the radioisotope to be removed in the same counter under the same conditions, but in a subtract mode, and observing complete subtraction on the oscilloscope display of the multichannel analyzer. This technique was soon extended to stripping by means of taped spectrums and a variable multiplier incorporated in the multichannel analyzer. Both magnetic-tape and punched paper-tape units became available. This technique is very useful for rapidly obtaining analytical results, even in the presence of overlapping photopeaks. The same technique can be used not only to remove interferences but also to compare directly the activity of an unknown with that of an activated standard of that element (or its tape-recorded spectrum). This can be done without computing photopeak heights or areas by using the counting time required with the standard, or the variable multiplier setting required with the standard, for complete subtraction of the photopeak of interest.

The spectrum-stripping method has two main disadvantages: its reliance on the oscilloscope display for visual estimation of complete subtraction and its sensitivity to gain shifts. For accurate work the total gain of the detector-analyzer system must be almost exactly the same for both sample and standard. Even a difference of one channel introduces uncertainty, and differences of only a few channels negate the method. Selecting photomultiplier tubes for minimum gain shift has been described by Covell and Euler[4] and Covell.[10,25] Fite et al.[4] described a drift-control device, and Guinn and Lasch[25] described methods of minimizing gain shift.

AUTOMATION AND COMPUTER TECHNIQUES

The instrumental method lends itself to automation and computer techniques. Kuykendall et al.[12] and Fite et al.[19] described automatic sample-handling and counting systems for such work. Guinn[52] described the application of automated systems to process control.

Computer techniques for the resolution of complex gamma-ray spectrums have been described by Nostrand et al.,[47] Kuykendall and Wainerdi,[47] Breen et al.,[53] Anders and Beamer,[54] Heath et al.,[55] Brice and Heath,[56] Burrus,[24] Morrison and Sunderman,[24] Hull and Gilmore,[10] Wainerdi et al.,[2] Nostrand and Rossi,[25] Trombka,[10,25] Burrus and Bogert,[25] Chester et al.,[25] Monahan et al.,[25] Drew et al.,[25] Nicholson et al.,[25] Scofield,[25] Blotcky et al.,[25] and Steele.[57] The methods of least squares, spectrum stripping, and iterative unfolding are used by one or more of these authors. Isenhour and Morrison[10] described a computer technique for the optimization of irradiation, decay, and counting conditions in activation analysis.

RELATED ACTIVATION-ANALYSIS TECHNIQUES

Although this paper is essentially limited to the field of neutron activation analysis, some mention should be made of recent advances in the closely related fields of charged-particle, photonuclear, and prompt-gamma activation analysis. The interesting possibilities of ^{3}He charged-particle activation analysis have been discussed by Mahony and Markowitz.[10,58] Photonuclear activation-analysis studies have been reported by Lukens et al.,[47,59] Otvos et al.,[60] and Lukens.[10] Photoneutron methods have been applied to the determination of deuterium by Guinn[8] and to the determination of beryllium by Guinn,[8] Goldstein,[61] Levine and Surls,[62] and Postma and McMurray.[19] Activation analysis using the prompt gamma rays emitted in either thermal-neutron capture or fast-neutron inelastic scattering has been described by Caldwell,[10] Greenwood et al.,[14,53] Greenwood and Reed,[4,10,24] Haffner et al.,[3] Clark and Rasmussen,[24] and Waggoner et al.[10]

New Applications

As the capabilities of neutron activation analysis have increased with the availability of better equipment and new techniques, the method

has found increased application in science and industry. Some of these new applications are described below.

PETROLEUM AND CHEMICAL INDUSTRIES

These industries have been leaders in the industrial application of neutron activation analysis, especially with the small neutron generators described earlier. Several companies use these generators for routine analyses at the 1- to 100-ppm and higher levels to determine nitrogen, oxygen, the halogens, and numerous metals in cores, crude oils, feedstocks, catalysts, polymers, and synthetic rubber. Relevant published papers include those by Anders,[10] Caldwell,[10] Caldwell and Mills,[63] Gray and Cartwright,[10] Guinn and Wagner,[34] Guinn et al.,[40,64] Iddings,[10] Johnson,[10] Mott and Rhodes,[10] and Steele.[2]

COAL INDUSTRY

Recently, conventional neutron activation analysis and prompt-gamma techniques employing a neutron generator have been applied to the rapid and routine analysis of coal for its carbon and ash contents (aluminum, silicon, and oxygen). Nuclear techniques in coal analysis were described by Martin et al.[10,24]

METAL AND SEMICONDUCTOR INDUSTRIES

With the growing importance of very pure metals (e.g., the alkali metals and beryllium) and semimetals (e.g., silicon and germanium) neutron activation analysis is finding increased application. Recent studies have been reported by Burns,[10] Guinn,[30,65] and McCrary et al.[4] The determination of oxygen in metals, especially, has attracted much attention.

PESTICIDE RESIDUES

The possible toxic effects to man and lower animals from wide-scale and excessive applications of pesticides in farming have become of great concern. In the determination of tolerance levels and violations of government regulations, large numbers of analyses for pesticide residues in crops and processed foods are required. Guinn and Potter[66] found that bromine residues in crops resulting from soil fumigation with bromide-containing nematocides could be rapidly determined by neutron activation analysis. As many as 240 samples can be activated in a single 30-min irradiation, and the induced 36-hr ^{82}Br activity can be rapidly determined instrumentally after 2 days' decay. Lindgren et al.[67] extended the technique to the determination of bromine residues in stored grain resulting from fumigations with methyl bromide. Castro and Schmitt[68] studied bromine residues from nematocides and also traces of DDT in plant extracts. Schmitt and Zweig[69] measured the DDT content of butterfat samples by the same technique. Guinn,[70] using neutron activation analysis, determined traces of mercury in wheat that resulted from illegal contamination with mercury-treated seed. Applications of the technique for the determination of pesticide residues have been summarized by Schmitt,[71] Buchanan and Guinn,[72] and Guinn and Schmitt.[73]

GEOLOGY AND GEOCHEMISTRY

All the rare-earth elements in meteorite and rock samples have been accurately determined by Schmitt et al.,[74,75] Mosen et al.,[76] Murthy and Schmitt,[77] Goldberg et al.,[78] Schmitt and Goles,[10] and Schmitt and Smith.[79] Corless and Winchester[10] found precise values for the calcium-isotope ratios in rocks by determining the ^{48}Ca by neutron activation analysis and the total calcium by chemical methods.

SCIENTIFIC CRIME DETECTION

One of the most fascinating new applications of the technique is in criminalistics. The ultrasensitivity of the method, which is achieved by use of the high neutron flux of a reactor, can extend the scope of the professional criminalist in his examination of physical evidence. Where the method can be applied in its purely instrumental form, the samples can be preserved for presentation in court.

General Atomic has developed a new activation-analysis technique for identification of gunshot residues on the gun hand of a suspect. The very unreliable chemical test (the paraffin test), which is no longer accepted in court, is based on the detection of nitro compounds removed from the back of the hand with paraffin. These compounds form a blue color with diphenylamine reagent. The activation method is based on the detection of traces of barium and antimony similarly removed from the hand. These elements are present in the primers of cartridges, and microgram or submicrogram

amounts are deposited on the hand when a revolver, automatic pistol, or rifle is fired. The amounts deposited by even a single firing are readily determined by 30-min activation in the reactor, radiochemical separation of 84-min ^{139}Ba and 2.8-day ^{122}Sb with carriers, and counting. Usually only slight traces of these two elements are found on the hands of persons who have not recently fired a gun. Deposits of barium and antimony are also found on the face of a person firing a rifle. Studies are now in progress in which trace levels of various high-sensitivity elements are incorporated in the powder of the cartridge. These elements are less common than barium or antimony, can be detected even more sensitively by neutron activation analysis than barium or antimony, and make it possible for various manufacturers to code their gunpowders with different tracer elements.

At GA many other materials of forensic interest have also been examined for their trace-element characterization possibilities. These include soil, paint, glass, paper, tire rubber, automobile grease, and hair. Activation analysis shows great promise for identifying the source of even a tiny sample of material evidence, and evidence from this method is now being introduced in court. Portions of this work have been published by Ruch et al.[10,80–82] and by Guinn.[83]

Forensic studies of paint, hair, opium, and other types of samples have been reported by Leddicotte et al.[80] and Bate et al.[19,80,84] Studies of human hair samples at GA and at Oak Ridge National Laboratory have substantiated and extended the findings on hair reported by Jervis in Canada and Lenihan in Scotland, i.e., that the trace-element composition of hair can provide truly individual characterization. Neutron-activation-analysis data were introduced for the first time into a U. S. court on Mar. 16, 1964, when Pro and Schlesinger presented analyses of soil samples involved in a liquor case. The data were accepted, and the case was won [see *Isotopes and Radiation Technology*, 1(4): 352 (Summer 1964)].

SPACE STUDIES

A highly miniaturized automatic neutron-activation-analysis device has been designed for remote, unmanned analysis of the surface of the moon in one of the forthcoming Surveyor missions. The device, weighing only about 30 lb, consists of a small sealed-tube accelerator source of 14-Mev neutrons, a scintillation counter, and a compact 256-channel pulse-height analyzer. Provisions have been made for activation with thermal and 14-Mev neutrons and measurement of both prompt and decay gammas. Various aspects of this program have been reported by Greenwood and Reed,[10,24] Haffner and Terrell,[24] Lee et al.,[80] Metzger,[10] Steele,[85] Trombka,[10] and Waggoner et al.[10]

OXYGEN DETERMINATIONS

The rather special technique of analyzing virtually any type of sample for trace and higher levels of oxygen by activation analysis with 14-Mev neutrons has found very wide application. Many laboratories in the United States now use a small neutron generator [10^{10} to 10^{11} n/(cm^2)(sec)] for rapid, routine, nondestructive determination of oxygen by the high-threshold (9.6-Mev) $^{16}O(n,p)^{16}N$ reaction. Since ^{16}N has a half-life of only 7.35 sec, irradiation and counting periods need only be 15 to 20 sec each. Samples are injected into the neutron flux, irradiated, and then ejected from the flux to the scintillation counter in a transit time of 1 to 2 sec. Since the gamma rays of ^{16}N are exceptionally energetic (largely 6.13 Mev), the determination is unique except in the presence of fluorine, which also forms ^{16}N by the $^{19}F(n,\alpha)^{16}N$ reaction. Fluorine, if present, can be detected and corrected for.

E. L. Steele at GA constructed an automated system in which there is a dual pneumatic-tube transfer system; sample and oxygen standard are rotated during activation, and the sample is counted with a pair of 3- by 3-in. NaI(Tl) crystals. The standard sample is counted by a single 3-in. crystal. Two single-channel pulse-height analyzers are used. Iddings[10] found the dual transfer system to provide better precision than can be attained with flux-monitoring systems. An electric motor rotates the sample and standard during irradiation, but in other respects the system is equivalent to the air-driven rotation device described by Anders.[10] With a fresh tritium target, samples can be exposed to 14-Mev neutron fluxes of about 10^9 n/(cm^2)(sec). Under these conditions the practical limit of detection for oxygen is about 0.1 mg, or 5 ppm in a 20-g sample. At higher oxygen levels precisions and absolute accuracies of ±2% of the value are attainable in a single determination. The same sample can be analyzed re-

peatedly, if desired, to provide better precision. A nitrogen atmosphere and special containers (copper, aluminum, and polyethylene) with low oxygen content are used to decrease the blank correction. Some samples are irradiated with no container. W. F. Harris at GA extensively compared oxygen values for various metals obtained by the activation method using 14-Mev neutrons with values obtained by the vacuum-fusion method. Agreement was excellent, even for oxygen levels down to 20 ppm. This comparison was reported by Guinn.[30] Lukens and Lasch[10] developed a Cerenkov counter for detection of the very-high-energy beta particles of ^{16}N (10.4 Mev E_{max}) even in the presence of very-high gamma-radiation levels, up to 1 r/hr.

Important papers on the fast-neutron method for determination of oxygen have been published by Anders,[10] Burns,[10] Iddings,[10] McCrary and Baggerly,[4] Steele and Meinke,[48] Veal and Cook,[49] and Volborth and Banta.[86]

PROCESS CONTROL AND USE OF STABLE TRACERS

The small neutron generator, coupled with the instrumental method, lends itself well to process-control applications. Several of these studies currently in progress in various industries were reported recently by Guinn.[52] Tarras and Pirtle[80] described an application of stable-element tracers, subsequently detected by neutron activation, in an air-pollution study.

Education

Four books on activation analysis have been published.[87–90] Literature reviews have been prepared by Meinke[91] for 1958–1959 and by Leddicotte[92,93] for 1960–1961 and 1962–1963. A 40-min professional-level training film entitled "Neutron Activation Analysis" has been completed for the U. S. Atomic Energy Commission by GA.

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Neutron Counting in Activation Analysis

By S. Amiel*

Paper 825 (abridged)

Neutron emission is a relatively rare phenomenon in radioactive decay. When it is observed under specified conditions or associated with another property (e.g., half-life), it can be used for identification and assay of specific nuclides and elements in an irradiated sample. The same is true for the emission of neutrons from a nuclear reacting system, especially with radioactive sources, where the emission is frequently characteristic of the source or target nuclides.

Neutrons can be detected selectively without any interference from beta- and gamma-ray background. This is an advantage compared with beta- or gamma-ray counting, where there is practically no specificity without chemical separation. Neutron detection also has advantages over gamma-ray spectrometry, where, without chemical separation, identification of characteristic peaks and their resolution might be very difficult in complex samples because of interferences from the multitude of peaks, shifts, and variations in the spectrums. Another favorable feature of neutron counting is the relatively high efficiency of detection with equipment that is simple and made of inexpensive components such as BF_3 proportional counters, water or paraffin moderators, and rather conventional electronic gear. The specificity of neutron emission and the salient features of neutron counting make neutrons a very useful analytical tool—analyses can be performed nondestructively and without radiochemical separations.

This paper reviews recent and current work at this laboratory on the development and use of neutron-counting techniques for analysis. The processes investigated for their analytical possibilities were delayed-neutron emission, photoneutron emission, and (α, n) reactions.

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Delayed-Neutron Emission

Except for isotopes of the heavy elements, which undergo spontaneous fission, the number of radioactive nuclides known to emit neutrons in decay is very limited, and their half-lives are short, ranging from a fraction of a second to some tens of seconds. These nuclides, known as delayed-neutron precursors, originate from nuclear reactions leading to neutron-rich nuclides having a few neutrons more than a closed-shell configuration. They decay by beta-ray emission to an excited state of the daughter, from which deexcitation by neutron emission is energetically possible. The neutron emission is an instantaneous process, but its rate is determined by the decay of the precursor, and it has the same half-life.

Most of the known delayed-neutron precursors are fission products; a few others have been observed in the products of high-energy spallation of medium-weight nuclei and in fast-neutron- or triton-induced reactions with some light nuclei. This review deals only with the observation of delayed neutrons from reactor irradiations, where the fission products* and ^{17}N are the only practical sources of delayed neutrons.

Delayed-neutron emission is quite rare, and the two sources have significantly different half-lives. Their production can be made specific by appropriate choice of source materials and irradiation conditions. The detection of delayed neutrons without interference is of great importance since an intense background is always present as a result of the radioactivation of other components of the sample.

FROM ^{17}N

The delayed-neutron precursor 4.1-sec ^{17}N can be produced directly by neutron irradiation in the reactions (Ref. 1)

$$^{17}O(n,p)^{17}N, \quad Q = -7.93 \text{ Mev} \qquad (1)$$

$$^{18}O(n,d)^{17}N, \quad Q = -13.77 \text{ Mev} \qquad (2)$$

or by secondary reactions with the 2.7-Mev tritons from $^{6}Li(n,\alpha)t$:

$$^{18}O(t,\alpha)^{17}N, \quad Q = 3.82 \text{ Mev} \qquad (3)$$

$$^{15}N(t,p)^{17}N, \quad Q = -0.15 \text{ Mev} \qquad (4)$$

The application of delayed-neutron counting for determination of oxygen by reactions 1 and 2 is possible only with high fluxes of fast neutrons; when ^{6}Li is present with oxygen, the production of ^{17}N from reaction 3 predominates. The very-high cross section of the reaction $^{6}Li(n,\alpha)t$ with thermal neutrons leads to a high yield of ^{17}N and makes it possible to use the delayed-neutron emission of ^{17}N for analysis. When nitrogen is present with ^{6}Li, ^{17}N can also be produced by the reaction $^{15}N(t,p)^{17}N$.

The extent of ^{17}N production depends on the degree of mixing of ^{6}Li with the oxygen- or nitrogen-containing medium because of the short range of the tritons. Solutions provide maximum intimacy and homogeneity and, when dilute, eliminate the need for corrections for self-shielding and attenuation of tritons. The production of ^{17}N in dilute ^{6}Li solutions irradiated with thermal neutrons is proportional[2,3] to the total ^{6}Li content of the sample and to the atom fraction of ^{18}O but independent of the water content. Hence the neutron emission from ^{17}N can be used to determine the ^{6}Li content of a solution if the atom fraction of ^{18}O of the sample is known; conversely, if the ^{6}Li content is known, the atom fraction of ^{18}O can be determined. Nitrogen-15 can be determined by an analogous process. The determination of ^{6}Li, ^{18}O, or ^{15}N by this technique need not be limited to solutions since any other intimate mixture of ^{6}Li and ^{18}O or ^{6}Li and ^{15}N will yield ^{17}N when irradiated with neutrons. The ^{17}N production parameters for solid mixtures, colloidal systems, or compounds should be determined specifically for each case since they depend on the physical properties of the sample.

A 20-sec irradiation results in the buildup of ^{17}N virtually to its saturation level, and counting its activity over an equal period is sufficient to record greater than 95% of its decay. Therefore analyses based on measuring ^{17}N are very rapid and require less than 45 sec for the entire procedure of irradiation and counting.

Determination of ^{18}O. The importance of ^{18}O as a stable tracer for oxygen has created the need for rapid analytical methods. The delayed-neutron count is directly proportional to the atom percentage of ^{18}O and is independent of sample size, provided the lithium content is fixed and the irradiation conditions are constant. Ratios of ^{18}O to ^{6}Li atoms ranging over many

*A section of this paper that discusses delayed-neutron emission from fission was not included in the *Isotopes and Radiation Technology* review.

orders of magnitude do not affect the proportionality between the neutron counts per unit weight of ^{6}Li and the ^{18}O concentration. This proportionality makes possible a selection of suitable ^{6}Li contents to give a neutron count in a predetermined range and thus decreases statistical errors in counting provided the ^{6}Li content is kept below a level where errors from self-shielding become important.

The reaction $^{18}O(t,\alpha)^{17}N$ may be used[3] for determining total oxygen. Intimate mixtures of the sample and ^{6}Li can be made by mixing fine particles in an inert-gas atmosphere, by fusing them together, or by dissolving them in an inert medium. The measurements are made in a way analogous to that in the determination of ^{18}O, and the total oxygen content may be derived if the isotopic composition of the oxygen is known.

Other Analytical Uses of ^{17}N. The formation of ^{17}N by fast-neutron irradiation of oxygen in the absence of ^{6}Li can form the basis for the determination of total oxygen in a sample. This can be done either with a high flux of fast neutrons in a reactor or with neutrons from an accelerator. A fission-spectrum neutron flux of 10^{14} n/(cm^2)(sec) will give at saturation a total neutron emission of about 650 n per milligram of oxygen.[1]

The production of ^{17}N by the reaction $^{15}N(t,p)^{17}N$ could form the basis for the determination of ^{15}N, which is an important stable tracer for nitrogen. Such an analysis requires ^{18}O to be absent from the sample or to be present only in known and very low amounts since its presence would contribute background that would have to be deducted from the measured count of the sample.

DELAYED-NEUTRON COUNTING

Analysis by counting delayed neutrons involves irradiation of the sample in a neutron flux from a reactor or from some other neutron source of sufficient strength, transfer to a neutron-counting assembly, and counting after a certain delay.

The counting apparatus used in conjunction with the Israeli Research Reactor-1 is outlined diagrammatically in Fig. 825-1. The assembly was a ring of six $^{10}BF_3$ neutron counters embedded in a block of paraffin. During counting the sample was situated in the center of the block. Pulses from the counters were fed

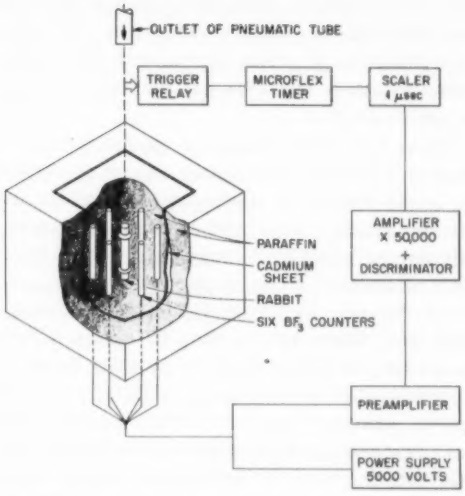

Fig. 825-1 Neutron-counting assembly and block diagram of electronic circuit.

through an amplifier to a fast scaler that was started automatically by an electronic timer after a definite preset delay period at the end of the irradiation. An array of borated-paraffin bricks around the counting assembly shielded the counters against background radiation.

Gamma-ray background was discriminated against efficiently, and the counting assembly results were reproducible over long periods. Photoneutron emission induced by other activities in the sample or sample container by the action of high-energy gamma rays on the deuterium of the paraffin was negligible.

The counting efficiency of delayed neutrons from fission and ^{17}N was about 10%.

Photoneutron Emission

Radioactivation analysis is commonly based on the formation of a specific radioactive nuclide and subsequent measurement of its radiations. When other activities are present and chemical treatment should be avoided or is impossible because of the short half-life of the nuclide in question, gamma-ray spectrometry is used to resolve the characteristic gamma ray of interest. When the resolution is insufficient because of a complex spectrum, coincidence analysis is practiced in favorable cases, e.g., gamma-gamma cascades. These techniques require relatively expensive instrumentation, and results are occasionally unsatisfactory. Destruction of the sample by chemi-

cal separation is then unavoidable; and, when the half-life of concern is very short, the concentration is at trace level, and the matrix is complex, then conventional activation analyses is unsatisfactory.

When the radionuclide in question has a relatively high-energy gamma ray, it is sufficient in most cases to discriminate against lower energy gamma rays and to measure the gross activity of radiation above this level. Further differentiation from other activities in the same energy range is made by following the half-life. When the nuclide of concern occurs in a complex matrix and its activity forms only a small part of the gross activity of a highly active sample, analysis without prior chemical processing is hindered by overloading of the detector and its paralysis during most of the counting time. Under these conditions electronic discrimination becomes difficult, and the accuracy and sensitivity decrease. Either chemical separations or, if nondestructive analysis is essential, a detector insensitive to gamma radiation below the energy in question is required to achieve the desired results. The photodisintegration of beryllium or deuterium and the emission of photoneutrons take place with gamma rays of energies greater than 1.67 and greater than 2.23 Mev, respectively. This phenomenon suggests that a detecting system based on counting the emission of photoneutrons from beryllium or deuterium would be entirely insensitive to energy quanta below the photoneutron threshold. An additional advantage of such a system would be the possibility of detecting emitted neutrons against any beta or gamma background.

The number of radionuclides that emit gamma rays with energies greater than 1.67 or greater than 2.23 Mev is relatively small, and their number can be decreased still further when a specific half-life range is selected. Thus a great degree of selectivity is indicated. This selectivity is the basis for the assay of nuclides emitting high-energy gamma rays and makes possible their determination by neutron or charged-particle radioactivation. The emission of photoneutrons from deuterium was used for the analysis of deuterium in water samples[4] by comparing their neutron activities with those of standards placed as targets around an intense source of ^{24}Na. Photoneutron emission from beryllium was used analogously for its assay. The method of analysis described here is the same in principle but has the roles of source and target interchanged; i.e., the source is unknown and the target constant as opposed to the beryllium or deuterium determination where the source is constant and the target unknown.

Clearly the form of the sample for such an analysis is important; it should not contain large quantities of neutron absorbers or material that could lead directly to neutron emission.

Samples containing large amounts of neutron absorbers lead to low values. Absorption is indicated when the photoneutron emission is not proportional to the sample weight. The true result can be obtained by adding a known amount of the element in question to the sample as an internal standard (spike). A sufficiently thick layer of beryllium or deuterium should be placed around the source to obtain the maximum output of photoneutrons. An efficient way to detect the photoneutrons is to count them after they have been thermalized by a moderator. Ideally, thermal-neutron counters should be placed in a 4π arrangement around the photoneutron source and immersed in a bath of heavy water or embedded in a beryllium block. Boron-10 trifluoride proportional counters are efficient neutron detectors of very-low sensitivity to gamma radiation. The irradiated sample is lowered into the counting assembly (Fig. 825-2) in the center of a flask containing 1 liter of heavy water (>99% D_2O). Neutrons generated in this container are counted with six $^{10}BF_3$ proportional neutron counters which are in a ring and are connected in parallel to a scaler and supplied from a common high-voltage source. The entire assembly is embedded in a block of paraffin, and external neutrons and gamma radiations are absorbed by a wall of lead and borated-paraffin bricks. The operating background is approximately 10 counts/min. Pulses from the counters are fed to a recording scaler that has its counting period controlled by an electric timer. Spurious counts from intense gamma radiation can be detected by replacing the heavy water with natural water and again determining the counting rate. The difference between the counting rates of heavy water and natural water gives the photoneutron counting rate of the source. The ratio of the counts from the two measurements agrees well with the ratio of their deuterium contents (1.5×10^{-4}).

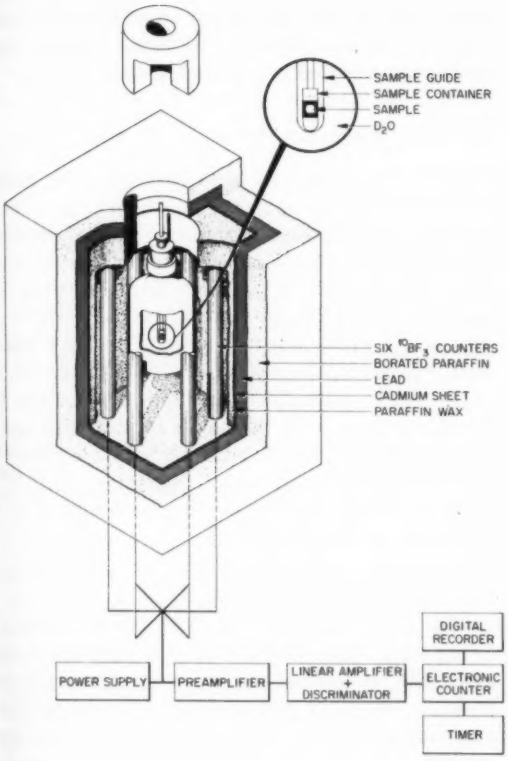

Fig. 825-2 Photoneutron measurement assembly and block diagram of electronic circuit.

Possible interference from sample constituents that lead to neutron emission was determined in the same way. In samples in which (α,n) reactions or fission could occur to give neutron counts not arising from deuterium, the interferences were detected and counted with natural water around the sample. Photoneutron emission from beryllium-containing samples was checked similarly with heavy water as the photoneutron radiator. When beryllium was used as a radiator, interference was checked by beryllium removal from or replacement in the assembly.

DETERMINATION OF SPECIFIC ELEMENTS

Sodium. Radioactivation analysis[5] of sodium is based on the formation of ^{24}Na by neutron capture. This isotope has a half-life of 15.0 hr and decays through a cascade of gamma rays of 2.75 and 1.37 Mev.

The photoneutron counting apparatus shown in Fig. 825-2 was calibrated and the precision of the method determined from the photoneutron emission rates of samples of known sodium content which were irradiated in various fluxes for varying times. No deviation from the 15.0-hr half-life was observed during an activity decrease of several orders of magnitude.

Absolute calibration of the apparatus was accomplished by observing the counting rate of photoneutron emission from standardized ^{24}Na sources. In the experimental apparatus described above, ^{24}Na gave 1 photoneutron count for every 2.45×10^4 disintegrations; the use of a larger D_2O container would decrease the ratio proportionally.

The photoneutron counting rate obtained from sodium after irradiation for 1 hr at a flux of 10^{13} n/(cm^2)(sec) was 15.6 ± 0.3 counts/min per microgram of sodium. The method permits, for example, the determination of 0.3 μg of sodium and has a precision of 10% when the sample is irradiated 1 hr at a flux of 10^{13} n/(cm^2)(sec). Higher sensitivities and better precision are possible with higher flux or longer irradiation.

A serious interference might arise from 14.1-hr ^{72}Ga, which at present cannot be distinguished from 15-hr ^{24}Na because of close relevant properties (photoneutron emission and half-life).

Manganese. Gamma rays of the high-energy part of the spectrum of 2.58-hr ^{56}Mn, in the same manner as those of ^{24}Na, produce photoneutrons from both beryllium and deuterium. This property was used[6] for nondestructive activation analysis of manganese and assay of ^{56}Mn by counting photoneutrons from heavy water. Although only 1 photoneutron for every 1.37×10^6 disintegrations of ^{56}Mn was observed under the experimental conditions, the detectability of manganese was 10 μg for a 1-min irradiation at a flux of 10^{13} n/(cm^2)(sec). The photoneutron activity observed was approximately 300 counts/min per milligram of manganese. The precision of the method is about 1%.

Sulfur and Calcium. Both sulfur and calcium are very difficult to determine by neutron activation and gamma-ray spectroscopy since the major radioactive products resulting from their irradiation are rather long lived and are pure beta emitters. Therefore chemical processing and beta counting are used.[7] Sulfur-37,

5.1-min half-life, produced from neutron capture in ^{36}S, and 8.8-min ^{49}Ca, produced from ^{48}Ca, are high-energy gamma-ray emitters; but, because of the very low abundance of their parent nuclides (0.014% ^{36}S and 0.18% ^{48}Ca), their assay by gamma-ray spectrometry, even with a multichannel analyzer, is normally very difficult because of summation peaks and other interferences that prevent their resolution in a highly active sample. Their short half-lives make chemical processing inconvenient. These difficulties are avoided by counting photoneutrons from D_2O in a method similar to that for sodium and manganese.

One neutron count was observed for approximately 1.1×10^4 disintegrations of ^{37}S or for approximately 1.7×10^4 disintegrations of ^{49}Ca. The specific activity measured experimentally was about 15.0 counts/min for 1 μg of sulfur irradiated 10 min in a flux of 10^{13} n/(cm^2)(sec), and the corresponding value for calcium was about 600 counts. The values were proportional over a wide range. The precision of the method for the determination of both sulfur and calcium was 2 to 3%.

Aluminum. Aluminum-28, 2.3-min half-life, is the only isotope of aluminum produced upon neutron capture. It is extremely difficult to separate chemically for subsequent activation analysis when present in a complex sample because of its short half-life. When present at trace levels, its 1.78-Mev gamma ray is frequently masked by the tail or sum peaks of the lower energy, but still very intense, gamma rays of accompanying activities. Gamma rays of 1.78 Mev are sufficient to cause photoneutron emission from beryllium. This property was used[8] for the determination of aluminum in alloys where, upon activation, the formation of gamma rays with energies equal to or greater than 1.67 Mev is rare. This phenomenon was verified by experiments that demonstrated the specificity of photoneutron counting for analysis of aluminum. The photoneutron activity observed was 430 counts/sec per milligram of aluminum irradiated 1 min at a flux of 10^{13} n/(cm^2)(sec). This value was proportional to the aluminum content. The precision of the method was about 2%, and 1 neutron count was observed for each 3.12×10^4 disintegrations of ^{28}Al.

Other Elements. The assay of ^{38}Cl for the determination of chlorine, of ^{140}La for lanthanum, of ^{72}Ga for gallium, of ^{151}Nd for neodymium, and of several other nuclides is being studied. Special attention is being given to fission products that might interfere.

Neutron Emission from (α,n) Reactions

Only the light elements interact with alpha particles to emit neutrons. With an appropriate sample matrix and a suitable alpha source, certain light elements present in the sample can be assayed, e.g., beryllium in ores by means of a polonium source; or the activity of a specific alpha emitter present and in contact with a given light element can be determined.

Of great importance is the interaction of alpha particles with the heavy isotope of oxygen. A very-high yield of neutrons was obtained[9,10] upon interaction of alpha particles with ^{18}O. This property was used for construction of a new type of neutron source.[11]

DETERMINATION OF ^{18}O

The determination[9] of the concentration of ^{18}O is very important in studying processes and reactions involving oxygen and in tracing its path in chemical and biological systems. The nuclear reaction $^{18}O(\alpha,n)^{21}Ne$ was used for analysis of various ^{18}O-labeled substances by bombarding the samples (in the form of gases, solids, or liquids) in an alpha-particle flux and then counting the neutron emission. The method is made feasible by the substantial yield of neu-

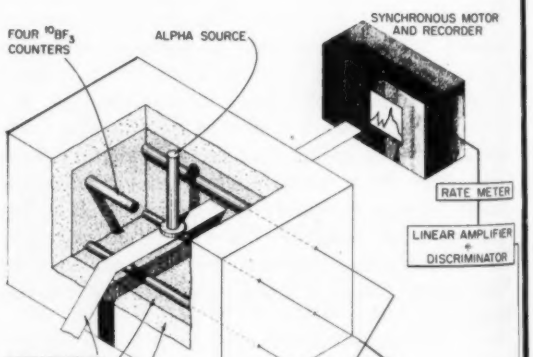

Fig. 825-3 Oxygen-18 paper chromatogram analyzer consisting of an alpha source and four neutron detectors embedded in a paraffin block.

trons from a thick ^{18}O target, e.g., 30 neutrons per 10^6 alpha particles from ^{228}Th and its decay products.[10]

An experimental assembly for scanning paper chromatograms is shown in Fig. 825-3. The

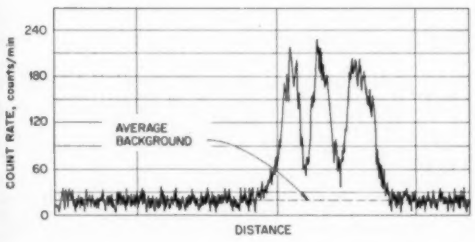

Fig. 825-4 Neutron-emission scan of a paper chromatogram containing three nearly identical bands of an ^{18}O-containing material.

efficiency was approximately 3% with four $^{10}BF_3$ neutron detectors. Sensitivities of a few micrograms of ^{18}O per square centimeter were obtained when fluxes of about 10^7 alpha particles/sec struck the paper chromatogram. The limiting factor was background from the structural oxygen of the paper. A typical scan of a paper chromatogram containing three nearly identical bands of an ^{18}O-containing material is given in Fig. 825-4.

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Determination of Trace Elements in the Greek Lakes by Neutron Activation Analysis

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Paper 854 (extracted)

In the past, spectrographic methods have usually been used for determining trace elements in lakes.[1] These and other conventional methods are, however, restricted for some elements because of insufficient sensitivity and large reagent blanks. One promising method of analysis for elements present in microgram or submicrogram quantities in lake waters is neutron activation analysis,[2-7] which has the advantages of great sensitivity[8,9] for many elements and elimination of reagent contamination and reagent blanks.

This paper reports the application of neutron activation analysis for the determination of 14 trace elements in the 11 most important lakes of Greece.

Experimental

Samples were collected with a Ruttner water sampler from the surface and at a depth of 5 or 15 m, depending on the maximum depth of the lake. The 14 elements investigated were silver, gold, arsenic, barium, bromine, chlorine, copper, iodine, manganese, potassium, sodium, rhenium, vanadium, and zinc.

Lake-water samples were filtered and pipetted into polyethylene tubes of 1.3-, 7.4-, or 30-ml capacity which were then heat-sealed. Volumes of standards equal to those of the samples were prepared as dilute aqueous solutions with triple-distilled water[10] and handled similarly. The concentration of a particular element in a standard solution ranged from 5.0 to 0.05 μg/ml and depended on the element under investigation and on the irradiation time. This range of concentration was selected to permit processing and counting after irradiation without additional dilution.

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To increase the sensitivity in samples analyzed for rhenium, vanadium, and zinc, 100 ml was evaporated to dryness and the residue was dissolved in 5 ml of distilled water and transferred to the polyethylene tubes for irradiation.

All neutron irradiations were done in the Democritus Reactor of the Nuclear Research Center of Greece. For irradiations up to 30 min, the pneumatic-transfer rabbit system was used. This system permits irradiation at a thermal-neutron flux of 2×10^{11} n/(cm^2)(sec) and delivery of the sample to the laboratory within 7 sec after irradiation. Irradiations with the rabbit system were made for chlorine, iodine, sodium, silver, and vanadium.

In-pool irradiations were made for the other elements under investigation. Irradiation times were 2 hr for barium, copper, and manganese and 8 hr for arsenic, bromine, gold, potassium, rhenium, and zinc. Samples and standards in the polyethylene tubes were suspended vertically in the reactor with nylon strings. The thermal-neutron flux at the sample location was about 2×10^{11} n/(cm^2)(sec).

INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

Excellent studies on instrumental or nondestructive activation analysis have appeared in the literature recently.[11–14] We applied this technique for chlorine, sodium, and silver.

Chlorine and Sodium. Samples and standards of sodium and chlorine were placed in the rabbit for 20-min irradiations. Aliquots of samples and standards were pipetted into culture tubes for counting, and the photopeak areas of ^{38}Cl and ^{24}Na were measured with a 3- by 3-in. NaI(Tl) scintillation crystal coupled with a 400-channel pulse-height analyzer. Memory data were recorded with a fast electric printer. Peak areas of samples and standards, with correction of the counting rates to the same decay time, were compared to determine these two elements.[15]

Silver. Half-milliliter lake-water samples were irradiated for 48 sec in the special polyethylene tubing[16] together with a weighed gold monitoring foil. Within 25 sec after irradiation, the induced activity of ^{110}Ag was counted for 24 sec, and data were transferred to a magnetic tape. Decay studies were made by counting the sample for 48-sec time intervals. Standards were counted in the same way. Gold foils weighing 0.5 mg were counted in a gamma scintillation counter for use in estimating the flux fluctuations, and all neutron irradiations were normalized to 2×10^{11} n/(cm^2)(sec). The ^{110}Ag was determined by comparison of the 0.66-Mev photopeak area of the sample with that of the standard.

CHEMICAL SEPARATIONS

Fast radiochemical separations[17,18] based on solvent-extraction techniques[19] were performed for vanadium, iodine, arsenic, rhenium, zinc, and gold.

Vanadium. The cupferron-chloroform extraction procedure described by Fukai and Meinke[20] was used. The rabbit system was used in irradiation of 2-ml samples, vanadium standards, and weighed gold monitoring foil. Vanadium carrier was added, vanadium was extracted as cupferrate into chloroform, and portions of the chloroform solution were counted. Decay studies were made by counting samples and standards for 4-min intervals for 25 min. Results were determined from a comparison of the 1.43-Mev photopeak areas of ^{52}V for both samples and standards. Gold foils were used to normalize all neutron irradiations to 2×10^{11} n/(cm^2)(sec).

Iodine. The rabbit system was also used in 30-min irradiations of iodine standards and lake-water samples. Gold solution was used for the estimation of flux fluctuations during irradiations. Iodine carrier was added, and samples were extracted into carbon tetrachloride by the procedure described by Glendenin and Metcalf.[21]

Silver iodide was precipitated, and the precipitates were filtered, washed, dried, and then counted in a well type scintillation counter. Gross decay was followed by gamma counting the samples for 25-min intervals for 2 hr. The radiochemical purity was checked with the 400-channel analyzer.

Barium and Manganese. Samples, together with barium and manganese standards, were irradiated in the reactor pool for 2 hr. Barium and manganese carriers were added as well as holdback carriers of arsenic, copper, sodium, and strontium. Barium and strontium were precipitated as nitrates, and the barium was processed by the procedure of Leddicotte.[22] The supernatant liquids from the barium and strontium precipitations were then processed for

manganese by another Leddicotte procedure.[23] Addition of perchloric acid, solid potassium chlorate, and concentrated nitric acid accelerates the precipitation of manganese dioxide.[24]

Arsenic. Arsenic was separated by an extraction procedure developed by Leddicotte and Grimanis.[25] More than 99.5% of the arsenic as As^{3+} is extracted into benzene in less than 2 min.

One milliliter of arsenic carrier solution and 1 ml of sodium holdback carrier solution were added to the irradiated samples. The reduced arsenic was extracted quantitatively from the solution into benzene and then stripped into distilled water. Extraction and stripping steps require less than 6 min. The As^{3+} was reduced to the metal in hydrochloric acid solution by 1 g of ammonium hypophosphite. After digestion and centrifugation the metal was filtered, washed, dried, weighed, and then counted in a well type scintillation counter. Decay was followed by counting ^{76}As at half-life intervals. The chemical yield was better than 90%, and no radiochemical contaminants were found.

Rhenium and Zinc. Rhenium and zinc were separated by an extraction procedure developed by Leddicotte and Grimanis.[26] Rhenium and zinc carrier solutions were added to the irradiated samples together with a saturated solution of hydroxylamine hydrochloride and a sodium holdback carrier. The solution was adjusted to 4*M* HCl, and the rhenium and zinc were extracted into tri-*n*-butyl phosphate (TBP) in benzene. More benzene was added to the organic phase, and the rhenium and zinc were stripped from the organic mixture into distilled water. Zinc was precipitated as ammonium phosphate,[27] and rhenium in the supernatant was precipitated as tetraphenylarsonium perrhenate.[28] The precipitates were filtered, washed with distilled water and ethanol, dried, and counted with a well type scintillation counter. The radiochemical purities of ^{69m}Zn and ^{186}Re were checked with the multichannel analyzer, and chemical yields were calculated. The separation time for rhenium and zinc was about 15 min.

Gold. Gold was extracted from hydrochloric acid solution into ethyl acetate by the procedure of Goldberg and Brown.[29] The only modification was the use of the method developed by Plantin[30] to strip the gold from the ethyl acetate into 8% NH_4OH.

Bromine, Copper, and Potassium. Bromine was precipitated as silver bromide. Counting of ^{82}Br was delayed a few hours to allow ^{38}Cl and ^{128}I to decay. Copper was precipitated as copper thiocyanate and potassium as perchlorate by procedures described by Leddicotte.[31,32]

Discussion

The flux of our reactor was too low to provide sufficient sensitivity for measuring rhenium and silver in the lake waters. However, the data obtained indicate that activation analysis should play an important role in supplying information on elements present in microgram or submicrogram quantities in lakes. Our results for some elements may be not only of geological importance but also of biological interest. The presence or absence of some trace elements may result in the eutrophic, oligotrophic, or atrophic behavior of the lake.

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Determination of Certain Trace Impurities in Pure Lead and Aluminum by Activation Analysis

By H. K. El-Shamy, A. A. Abdel-Rassoul, and T. Z. Bishay*

Paper 829 (abstracted)

The presence of microquantities of certain metallic impurities in lead usually limits its industrial application, and even traces of foreign elements in aluminum may prevent its use as reactor material. The conventional analytical methods for both metals have numerous and substantial limitations. Activation analysis may be used with considerable accuracy for determination of several elements in different materials, particularly elements present in concentrations far below those determined by other microanalytical techniques. Determination of impurities in lead and aluminum by activation analysis has been described by various authors.[1–5]

In this paper a systematic determination of several elements (Cr, Co, Sn, Fe, Zn, and Cd) present as trace impurities in lead and aluminum samples of different purity grades is described. A separation scheme based on coprecipitation and ion-exchange techniques was developed (Fig. 829-1). Samples and standards were irradiated 48 hr at a flux of 1.3×10^{13} n/(cm^2)(sec) in the UA-RR-1, the 2-Mw research reactor at Inshas. After decay for 7 days, samples and standards were dissolved in appropriate acids, and carrier solutions were added. Macroquantities of both lead and aluminum, as $PbCl_2$ and $AlCl_3$, were readily separated from a cooled (0°C) sample solution exposed to the prolonged action of hydrogen chloride gas. The chlorides soluble in concentrated hydrochloric acid were subsequently passed through a column of a strongly basic anion exchanger, Dowex 1, and the metallic ions retained were successfully fractionated with eluant of various concentrations of hydrochlo-

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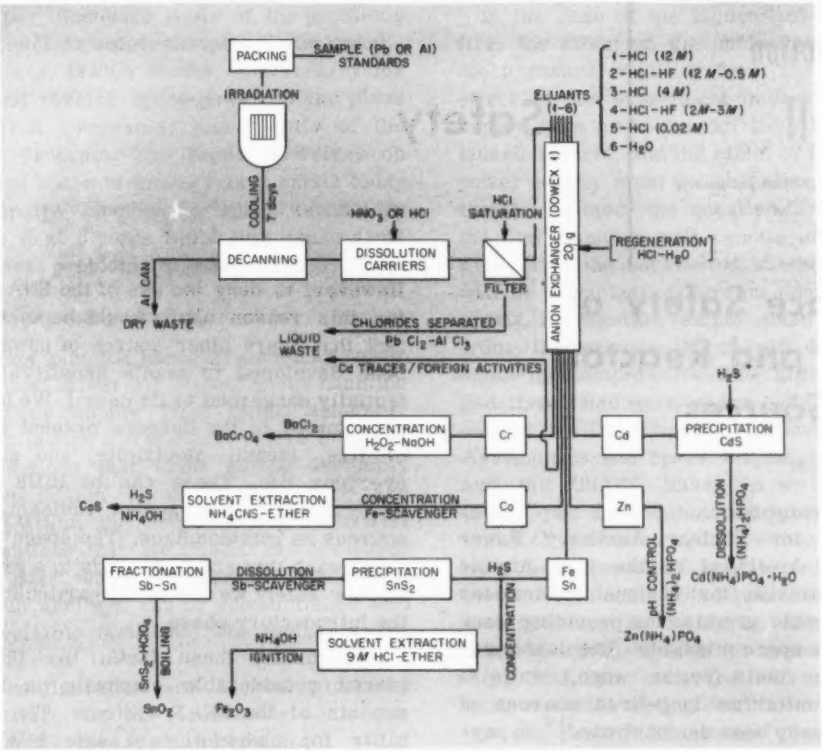

Fig. 829-1 Schematic diagram for chemical separation of elements determined in lead and aluminum samples.

ric acid and hydrofluoric acid. The radionuclides of concern collected in the individual fractions were further purified by chemical precipitation before being compared with irradiated standards processed similarly.

Addition of carriers, rather than fractionation in the carrier-free state, enhances the purification of the radionuclides. Successive ion-exchange chemical-precipitation techniques ensure complete decontamination from foreign activities.

Most of the activity measurements were made with a shielded G-M counter having a mica-end-window thickness of less than 2 mg/cm^2 and a Philips scaler. The activities in the samples and their respective standards were determined under identical geometries. Whenever possible, decay curves were also determined to ensure the absence of short-lived activities, and aluminum absorption curves of the standards and samples were prepared and used to check the identity of the isotope counted. The mean deviation determined from duplicates was ± 10 to 12%; however, the sensitivity can be appreciably increased by longer irradiation times, higher fluxes, and increased sample weights.

Gamma-ray spectrometry for the quantitative determination of the elements concerned[5] was tried, but the low counting rate of the samples investigated limited its application. The present technique is satisfactory, and no interference of foreign activities was detected. The method described is now used for routine analysis of both metals with good reproducibility.

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Section

II

Isotopes and Radiation Technology

Safety

Aerospace Safety of Isotopic and Reactor Power Sources

By Glenn A. Fowler*

Paper 286 (abridged)

The Systems for Nuclear Auxiliary Power (SNAP) being developed by the U. S. Atomic Energy Commission for various applications show considerable promise for providing electric power for space missions. The usefulness of radioisotope units (power range,† watts to hundreds of watts) as long-lived sources of power has already been demonstrated[1,2] in payloads placed in orbit in 1961 and 1963. For applications where higher power is demanded, reactor units (power range,† hundreds to millions of watts) offer promise. Although no reactor unit has yet been flown, the USAEC has stated its interest in seeing development carried through the demonstration phase, including both ground and flight testing.

The SNAP systems derive their power from the heat generated by the spontaneous nuclear decay of radioisotopes or from the controlled nuclear fission in a reactor. Since the basic origin of the energy is the nuclei of the atoms of the source material, it is independent of chemicals, sunlight, magnetic fields, etc. The SNAP systems can provide reliable power for relatively long times; this characteristic makes them ideal for space missions. Their principal disadvantage is that these radioactive materials are potentially dangerous to human life. However, to deny the use of the SNAP systems for this reason alone would be to ignore the fact that every other source of power that has been developed to man's benefit also is potentially dangerous to its user.‡ We have grown accustomed to the dangers present in our use of fire, steam, electricity, and gasoline in everyday life. There can be little doubt that we will ultimately accept radioactive power sources as commonplace. The speed with which they reach this status depends to a great extent on how safely we use them, particularly during the introductory phase.

Recognizing these facts, the USAEC has placed considerable emphasis on the safety aspects of the SNAP systems. The responsibility for providing a safe SNAP system throughout its factory-to-orbit sequence is placed squarely on the shoulders of the power-supply designer. In addition, the Assistant Director for Nuclear Safety, Division of Reactor Development, USAEC, makes an independent assessment of the adequacy of the safety plan for each SNAP system. This assessment involves a complete study of the safety plans for the system as well as its proposed application to a space vehicle. It requires thorough analytical studies and may involve both ground and flight testing. Sandia Corporation is the principal USAEC contractor for the aerospace nuclear safety program.

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†Editors' Note: Since this paper was written, values have changed. Radioisotope units in the multi-kilowatt range are now envisioned.

‡Paper 288, Atomic Energy for Society and the Balance Between Hazard and Gain, by J. H. Sterner was printed in *Nuclear Safety*, 6(2):143-145 (Winter 1964–1965). Sterner points out the necessity for intelligent evaluation of the relation of benefits to hazards, for which a reasonable set of guides has not yet been established. He further emphasizes that, in the nuclear industry, control of the health aspects of radiation is already far ahead of other environmental health problems. [See also *Isotopes and Radiation Technology*, 2(2): 172-181 (Winter 1964–1965).]

This paper discusses some of the problems related to nuclear safety associated with launch-pad accidents, launch aborts, and reentry for isotopic and reactor space power plants; plans for research programs; and results of the flight-test program. The paper is written on the basis of space technology as it exists today but anticipates changes in space technology during the next 5 years which may cause some of the present problems to disappear.

Factory-to-Reentry Safety

The safety plan for aerospace nuclear power supplies must cover all phases of the handling of the power supply from initial assembly through its launch and possible return to earth. Safety questions that arise during assembly and transportation to the launch area are generally within the scope of the industrial safety practices that the USAEC has developed over the past years. The testing required to simulate an accident can be accomplished with readily available facilities. The nuclear power supply, whether isotopic or reactor, is in a shipping configuration and is well protected against failure for all foreseeable accidents, including shock, fire, and immersion.

However, when the flight payload, including the nuclear power supply, has been mated to the launch vehicle and the launch countdown is begun, conditions are somewhat changed. The nuclear power supply no longer has the protection of its shipping configuration and is on top of a tall missile-shaped pile of explosive. At this stage five major possibilities attract the attention of the safety analyst: (1) the launch vehicle may blow up and burn on the pad; (2) it may lift off successfully but blow up or be destroyed while still in the general vicinity of the launch area; (3) it may fail to boost the payload to orbital velocity; (4) it may achieve a short-lived orbit with the payload returning to the earth in days or months; or (5) it may achieve a good orbit with the payload orbiting for years. In cases 1, 2, and 3, an isotopic power supply will be at its highest activity level; in case 4 it will be partially decayed; and in case 5 it will have decayed to a relatively low level. A reactor power supply will be at a low level for all cases except 5, assuming the reactor will be started only after it has been placed in a good orbit.

In the case of the launch-pad explosion and fire, the effect on the nuclear power supply of the pressure resulting from the explosion, the effect of the subsequent impact as the power supply falls or is thrown from the nose of the launch vehicle, and the effect of fuel fire on the power supply must be considered. If the system is isotopic, the question is whether or not the fuel capsules will remain integral; if it is a reactor, the question is whether or not there will be a nuclear excursion upon impact. Only spotty information exists about the pressure from the various launch-pad explosions and about the temperature-time history of launch-pad fires. Recognizing the need for this information, the Department of Defense, the National Aeronautics and Space Administration (NASA), and the USAEC have set up a coordinated launch-pad instrumentation program. Various types of active and passive measuring devices have been installed at selected launch sites; they accommodate both liquid- and solid-propellant boosters so as to be able to fill in the missing information if an accident should occur.[3] The data collected will allow realistic environmental testing conditions to be set without the penalty of severe overtesting or the false security of undertesting. Since boosters do not blow up on the pad as frequently as they have in the past, information is coming in very slowly from this program.

Although the USAEC is focusing its attention primarily on the SNAP system design, the Commission also must concern itself with the manner in which the SNAP system is physically incorporated into the space vehicle. The reentry heating situation for a nuclear power supply buried inside a large flight vehicle obviously differs from that for a similar power supply mounted on the end of a boom and essentially independent of the flight vehicle during reentry.

If the missile leaves the launch pad successfully but fails or is destroyed early in flight, then the main sources of damage to the nuclear power supply are pressure from the explosion and the consequences of impact on water, soil, or surface structures. As in the case of the launch-pad accident, the isotopic supply must be designed to remain integral, whereas the reactor supply must be designed to prevent criticality under these circumstances. Fire is

less of a concern here, but greater velocities than in a launch-pad accident would produce more severe impacts. Furthermore, the impact area in this case will not be as well controlled from public access as is the launch pad.

The vehicle that falls just short of orbital velocity encounters essentially the same reentry conditions as those encountered in orbital decay; however, a significant difference is that the possible impact areas can be fairly well determined in advance. These potential impact areas can be kept over relatively unpopulated sea and land masses to minimize hazard. Also, an active control system aboard the flight vehicle can command actions relating to the nuclear power supply. For example, disassembly can be caused by mechanical or explosive means or the power supply can be cut loose from the flight vehicle.

The short-lived orbit (days to months) still offers the possibility of active control of safety actions but leaves the reentry and impact locations to chance. The long-lived orbit (years) denies reliable active control (with present technology) and denies identification of reentry and impact location, but it does offer time enough for the radioactivity to decay to a low level before reentry.

Reentry Burnup

For the last three of the five cases discussed—suborbital failure, short-lived orbit, and long-lived orbit—the flight vehicle and the nuclear power supply will be subjected to severe reentry heating conditions. The designer has the choice of designing the source for intact survival at the reentry temperature or of using the reentry heat pulse to cause complete burnup and dispersal in the upper atmosphere.

Intact reentry is attractive, particularly if the power supply could be controlled to impact in a designated area, because it essentially eliminates the major questions about public safety in the use of nuclear power supplies in space. If controlled impact could be easily achieved, the power supply could be directed to one of the existing land ranges and accurately tracked to impact. A recovery party could then go to the area and recover and dispose of whatever radioactive material remained. Unfortunately the instrumentation for controlled reentry adds weight that must be taken from the payload, i.e., the nuclear power supply. In addition, controlled intact reentry calls for a reliable active command and retro-rocket system aboard the space vehicle. Today's technology does not offer much assurance that such a system would be available in the long-lived-orbit case; however, it could be devised with high confidence for the short-lived orbit. As more experience with reliable electronics and with propellant life in space is gained and as other improvements in technology are made, reliability limitations probably will disappear, leaving only the extra weight as the price for controlled intact reentry. Another possibility for disposal would be to move the space vehicle and nuclear power supply to a long-time "junk" orbit where they would remain for nundreds of years.

In each nuclear power supply being considered so far for orbital flight, the designer has chosen to use reentry burnup and dispersal in the upper atmosphere as the method for disposing of the radioactive material. This choice has caused the aerospace nuclear safety program to investigate high-temperature-material properties, ablation droplet formation, and upper-atmosphere circulation patterns.

The problem of a material's surviving reentry temperatures, both on the relatively steep reentry trajectories of the ballistic missile and on the shallower reentry trajectories of the manned orbital flights, has received expert attention over the past several years. On the other hand, the deliberate use of orbital reentry heating for burnup and dispersal of various materials in assorted configurations has been of limited interest except to the designers of aerospace nuclear power supplies. As a result of the missile effort, the heating rate on simple shapes for a given set of reentry conditions can be calculated with high confidence by any one of several methods.[4] However, the irregular shape of the nuclear power supply (compared to nose cones) and the variety of materials incorporated in the typical nuclear power source present some interesting problems to the designer and the safety analyst. To determine the expected heat input on various parts of a complex geometry, hypervelocity or hypersonic shock tunnels, such as those operated by the Arnold Engineering Development Center of the Air Force or the Cornell Aero-

nautical Laboratory, Inc., must be used. In such facilities it is possible by full- or fractional-scale-model tests to establish the heat-input relations between the simple shapes for which computational models exist and various parts of the more complex geometry of the specific nuclear power supply being examined. With these inputs applied to a thermal model of the nuclear power supply, if the properties of the materials are known, it is possible to calculate the time of melting and disassembly of the various parts of the power supply when it is exposed directly to the reentry heating pulse.

Although shock tunnels can be used to provide short runs at high heat-transfer rates, there is at present no facility that can realistically reproduce reentry flight conditions for times comparable to those of actual orbital reentry. Such a facility would allow rapid determination of the effect that a design change in the nuclear power supply would have on its burnup characteristics and would allow experiments such as the measurement of ablation droplet-size distribution. It would be a powerful tool for the study of any reentry heating phenomena. Unfortunately there is no immediate prospect for such a facility. Judging from experience with the development of other environmental facilities, it is likely that any practical reentry heating facility will only partially simulate the actual reentry conditions and that dependence on analytical methods and on actual flight testing will continue to be heavy.

Research Program Plans

The first step in an assessment of the efficiency of the reentry burnup and dispersal characteristics of a nuclear power supply in a particular application is an examination of the physical arrangement for its incorporation into the flight vehicle. The aerodynamic characteristics of the flight vehicle during reentry must be determined so that the vehicle attitude will be known and hence the nature of the reentry heating to which the power supply will be subjected. A good vehicle design will place a SNAP device designed for burnup in a position to be exposed to the full effect of the aerodynamic heating throughout reentry.

Once the character of the reentry heating has been established, the power supply is "flown" in the computers by the Aerospace Nuclear Safety Organization to determine whether or not the heat available from reentry is sufficient to melt the container and allow ablation of the radioactive material. The lack of complete data on properties of materials at and above the melting point has caused the Aerospace Nuclear Safety Organization to ask Oak Ridge National Laboratory to compile available information on thermal and chemical properties of various materials and to evaluate our country's capability of providing the needed information.

If reentry heating calculations fail to show complete melting of the radioactive material in the nuclear power supply during reentry, then burnup and dispersal will not be accomplished. However, when melting of the fuel is calculated to occur, then the size of the particles that will result and where they will be distributed in the upper atmosphere become of interest. This is probably the area of greatest uncertainty in reentry burnup studies, and steps are being taken to develop methods of determining particle-size distributions under various conditions.[5] Zirconium metal has shown the property of exploding promptly into many very small particles when molten droplets of a few hundred microns in diameter are exposed to the atmosphere. It is not yet known how many other materials will show similar properties or what is the minimum size of droplet that will have this explosive property. The Illinois Institute of Technology Research Institute is studying and evaluating the possible coalescence of small-particle ablation debris to form larger masses of material.[6]

Even when particle size and distribution in the upper atmosphere resulting from reentry burnup are known, uncertainty as to when and where the material will reach the biosphere still remains. Although the general behavior is known for various ranges of particle sizes and altitudes, additional studies have been undertaken by the Geophysics Corporation of America (GCA) and by Travelers Research Corporation (TRC). The GCA is striving to derive an analytical model that will indicate the fall and dispersion of particles that are injected at an altitude of 250,000 ft and fall to 10,000 ft. The TRC is concerned with dispersion at 100,000 ft and below. Both efforts are primarily theoretical but could lead to development of an

experimental program if existing data prove inadequate.

Reentry Flight Testing

Flight testing is an important phase of the nuclear safety program but is expensive and can provide only relatively few data per flight. After analytical studies, computations, and laboratory testing have been used to the fullest in the assessment of the safety features, flight testing demonstrates the safety characteristics of the nuclear power supply under actual reentry conditions.

The first Reentry Flight Demonstration (RFD-1) in this program took place[7] in May 1963. This was an attempt to verify the operation of design features intended to enhance disassembly of the reactor under reentry heating conditions, to verify temperature-rise calculations at various points in the reactor mock-up during reentry, and to obtain gross indications of the rate of burnup of full-scale models of the reactor fuel rods that were independently exposed to the reentry heat pulse by ejecting them from the reentry vehicle.

The reentry vehicle carried a full-scale instrumented mock-up of the reactor assembly and was equipped with a C-band radar beacon, an FM-FM radiotelemetry system, and a 100-sec-delay tape recorder designed to permit data transmission following emergence of the telemetry signal from the ionization blackout caused by the reentry heating. Data were transmitted continuously both in real time and with the 100-sec delay. The reactor mock-up was instrumented with thermocouples for temperature-rise measurement and with action switches that would show when physical parts were removed from the mock-up as a result of reentry heating. The reentry vehicle contained a parachute and recovery aids so that, if it survived the reentry, it might be found and examined. The vehicle was placed under the desired reentry conditions (−6°, 20,000 ft/sec, 400,000-ft altitude) by a NASA Scout vehicle. This four-stage solid-propellant system was launched from Wallops Island, Va., and placed the reentry vehicle almost perfectly on the flat trajectory required to bring it into range of the instrumentation.

Radiotelemetry from the transmitter aboard the reentry vehicle was received and recorded at Wallops Island, at the NASA Mercury station on Bermuda, and in the downrange area aboard aircraft operated by the Air Force Missile Test Center, the Air Force Special Weapons Center, and NASA. Good radiotelemetry signals were received from lift-off until ionization blackout of the signal occurred 350 sec later at an altitude of 200,000 ft. No signal appeared after the blackout effect was over. It is believed that severe reentry-vehicle damage occurred during the blackout period when the peak heating rate was encountered. The C-band beacon mounted in the reentry vehicle was tracked by NASA instrumentation radar at Wallops Island and at Bermuda. Tracking was excellent, and trajectory information was good until the ionization blackout blocked the radar-beacon transmission.

Optical observations were made by Sandia Corporation equipment on Bermuda. Tracking signals from the NASA FPS-16 radar on Bermuda were used to point two long-focal-length tracking telescopes in the right direction until the operator was able to sight the vehicle and track it manually. Fixed-plate cameras were mounted to cover the visible portion of the trajectory. Spectrographic and photometric coverage also was provided. Several of the aircraft carried cameras for photographing the reentry. Unfortunately the light level received at the Bermuda station was below even the most conservative estimates; thus the tracking telescope and the photometer film were useful only during the brightest part of the reentry. Records from the fixed-plate cameras and from the spectrographic instruments on Bermuda and aboard some of the aircraft were good. The NASA aircraft that was in a favorable position obtained a very useful movie film that covered the final part of the trajectory, as well as good spectrographic records.

The flight demonstrated that the disassembly features in the reactor design worked properly. It also verified the supposition that temperature rises caused by reentry heating of the reactor model were predictable with reasonable accuracy and gave a gross indication of the burnup rate of the reactor fuel rods released from the reentry vehicle shortly after fourth-stage ejection. Subsequent testing and analysis on the reentry-vehicle design indicate that there were areas of more severe localized heating on the vehicle than originally anticipated. It is believed that the ablative covering was inadequate in some areas, and so burnthrough occurred. It appears that the irregular

shape of the reactor mock-up carried on the front of the reentry vehicle was a major contributor to the vortexes that caused the excessive local heating.

A second Reentry Flight Demonstration (RFD-2), planned for the fall of 1964,* will carry a model of an isotopic power supply instrumented to investigate experimentally the disassembly features incorporated in the power-supply design and to provide information on fuel-capsule ablation by techniques similar to those used on RFD-1 for the reactor fuel rods. The instrumentation aboard the reentry vehicle and at the ground station is similar to that used for RFD-1 but with increased emphasis on optical coverage. The vehicle was designed with increased protection and better experimental determination of the reentry heat-load distribution. It is expected to survive reentry and to be recovered from the Atlantic Ocean about 200 miles southeast of Bermuda. Recovery will provide information about the burnup of the nuclear-power-supply model and about the reentry-vehicle design parameters.

In both demonstration flights the information obtained on heating and burnup of the nuclear-power-supply model is limited to disassembly information. No information has been, or will be, obtained from these flights on the final particle size and distribution in the upper atmosphere because no practical method has yet been developed for obtaining such information during a flight test. Since this information would be an important input to the aerospace nuclear safety program, sampling by aircraft and rocket techniques is still under study with the hope that a practical system can be found. The possibility of tracking the debris cloud by radar or laser-radar techniques is being examined by Sandia Corporation. Knowledge of the dispersion of the debris cloud as a function of time could yield very useful information if a practical tracking scheme can be found.

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*Editors' Note: Paper was written before test, which took place Oct. 9, 1964. [See *Isotopes and Radiation Technology*, 2(2): 192 (Winter 1964–1965).]

†Editors' Note: References 1 and 2 were summarized in *Isotopes Radiation and Technology*, 1(4): 325-339 (1964).

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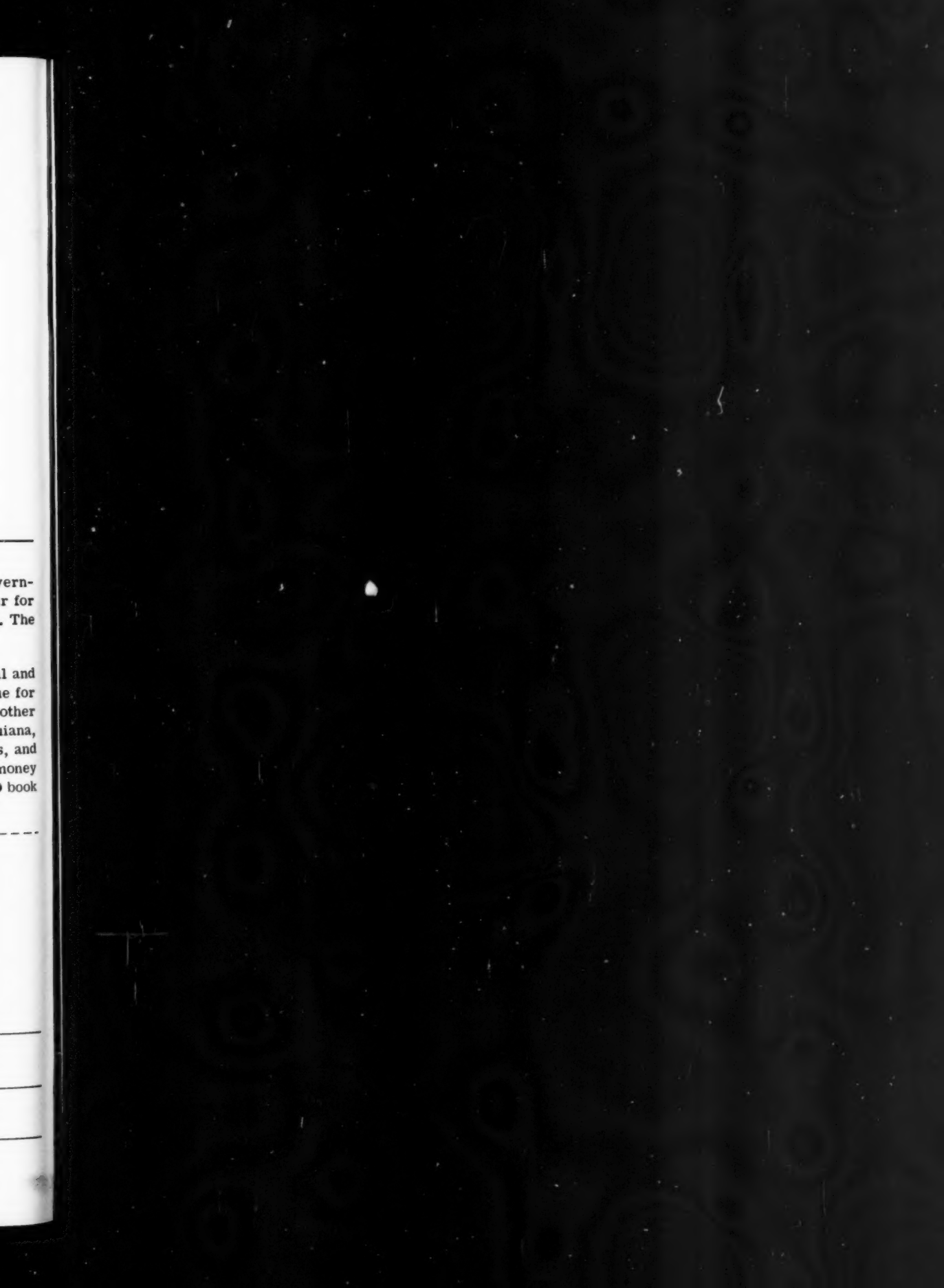
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